

# Eco-friendly gas insulating medium for next-generation SF<sub>6</sub>-free equipment

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## ABSTRACT

Gas-insulated equipment (GIE) that utilizes the most potent greenhouse gas sulfur hexafluoride (SF<sub>6</sub>) as insulation and arc-quenching medium has been widely used in the power industry. Seeking eco-friendly insulating gas with advanced performance for next-generation SF<sub>6</sub>-free GIE is significant for the “net-zero” goal and sustainable development. In this paper, the utilization, emission, and reduction policies of SF<sub>6</sub> around the world were summarized first. Then, we systematically reviewed the latest progress in comprehensive performance evaluation of eco-friendly insulating gas in terms of molecular design, dielectric insulation, arc-quenching, stability and decomposition, materials compatibility, biosafety, etc. Further, the representative applications of eco-friendly insulating gas in medium-voltage, high-voltage GIE as well as relevant maintenance-related technologies were highlighted. Accordingly, the existing challenges and future perspectives were proposed, presenting a roadmap to hopefully steer the development of eco-friendly insulating gas and GIE.

## KEYWORDS

Eco-friendly insulating gas, net-zero, dielectric insulation, arc-quenching, SF<sub>6</sub>-free gas-insulated equipment (GIE).

Gas-insulated equipment (GIE) including gas-insulated switchgear (GIS), gas-insulated transmission line (GIL), gas-insulated transformer (GIT), gas-insulated cabinet (GIC) with the advantages of small footprint, high reliability, long maintenance cycle has been widely used in the power system of various voltage levels<sup>[1,2]</sup>. Normally, sulfur hexafluoride (SF<sub>6</sub>) is utilized as the insulation and arc-quenching medium for GIE owing to its superior dielectric performance. However, SF<sub>6</sub> is one of the most potent greenhouse gases with a global warming potential (GWP) of 25,200 and an atmospheric lifetime of 3,200 years<sup>[3,4]</sup>. The power industry accounts for 80% of the SF<sub>6</sub> consumption, which value reaches over 7,000 tons in China<sup>[5,6]</sup>. Moreover, the global SF<sub>6</sub> emission amount demonstrates an increasing trend at the rate of 10% annually, whose atmospheric content changes from 3.67 part per trillion (ppt) in 1994 to 10.41 ppt in 2020<sup>[7,8]</sup>. The design and application of eco-friendly insulating gas to substitute SF<sub>6</sub> for next-generation GIE are necessary and urgent currently to achieve the “carbon peak, carbon neutral” and “net-zero” emission<sup>[9]</sup>.

Up to now, various gases have been focused as potential substitutes to SF<sub>6</sub> owing to their low GWP and superior dielectric strength, such as perfluorocarbons (PFCs), trifluoroiodomethane, fluorinated nitrile, fluorinated ketones, hydrofluoro-olefins (HFOs), etc. The assessment of the application feasibility of the proposed eco-friendly gas is also conducted to provide basic information for

GIE design and engineering application. Meanwhile, various theoretical analysis technologies have been relatively mature in exploring the micro-mechanism of gas insulation, leading to performance prediction and molecular design. Further, some pioneering eco-friendly insulating gas-based equipment has been developed for engineering applications. Relevant progress has also been summarized. Specifically, Rabie et al. and Franck et al. provided an overview of gas insulation, the climate impact of SF<sub>6</sub> and development of eco-friendly insulating gas for SF<sub>6</sub>-free medium-voltage (MV) and high-voltage (HV) GIE<sup>[10,11]</sup>. The advances in the quest for eco-friendly insulating gas were reviewed by Beroual et al.<sup>[12]</sup> Further, Li et al. and Zhang et al. summarized the fundamental physicochemical properties of eco-friendly insulating gas for GIS application, where the basic data calculations and fundamental experiments for performance evaluation were emphasized<sup>[13,14]</sup>. The stability and decomposition characteristics were reviewed by Xiao et al. and Yang et al, which guides the reliable application of eco-friendly insulating gas<sup>[15,16]</sup>. The in-detail characteristics of C<sub>4</sub>F<sub>7</sub>N, C<sub>5</sub>F<sub>10</sub>O were also summarized by Pan et al. and Owens et al.<sup>[17,18]</sup> Challenges primarily remained on the problems such as inferior stability, weaker arc extinguishing performance, hazardous by-product generation, etc. The feasibility of eco-friendly insulating gas for GIL application was also evaluated by Tu et al.<sup>[19,20]</sup>

Herein, we highlight recent advances in eco-friendly insulating gas for next-generation SF<sub>6</sub>-free GIE. Firstly, we provide an

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overview of the SF<sub>6</sub>-based GIE, followed by the emission and reduction policies of SF<sub>6</sub> to clarify the necessity of seeking eco-friendly insulating gas. Secondly, the basic requirements, categories and molecular design of eco-friendly insulating gas are summarized, indicating the main performance assessment dimensions. Thirdly, we highlight recent progress of various eco-friendly insulating gas in terms of molecular design, dielectric insulation, arc-quenching, stability and decomposition, materials compatibility, biosafety. After that, we summarized the latest application of eco-friendly insulating gas in MV, HV scenarios as well as relevant maintenance-related technologies. Finally, we discuss the existing challenges and propose perspectives for future research. We believe this review could provide insights into the development of eco-friendly gas insulating medium as well as next-generation SF<sub>6</sub>-free GIE.

## 1 Utilization and emission of SF<sub>6</sub>

### 1.1 SF<sub>6</sub>-based GIE

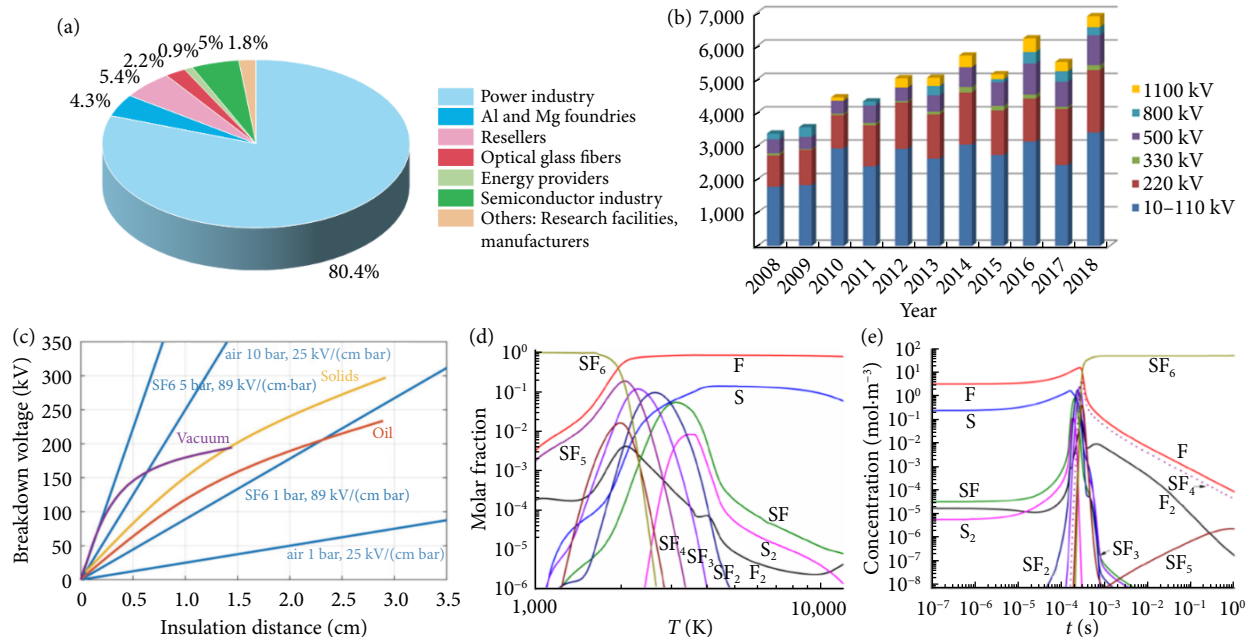
SF<sub>6</sub> is successfully synthesized by Moissan and Lebeau in 1900 and applied as dielectric insulation and arc-quenching medium since the 1960s<sup>[21]</sup>. Pure SF<sub>6</sub> is non-toxic, non-flammable, noncorrosive and physiochemical stable, which is suitable for various application fields including the power industry, mining engineering, semiconductor processing, optical engineering and health care<sup>[21]</sup>. GIE for electricity transmission and distribution accounts for 80% of SF<sub>6</sub> consumption, followed by magnesium casting and aluminum smelting (as shown in Figure 1(a)). Specifically, the annual SF<sub>6</sub> utilization in China demonstrates an increasing trend (Figure 1(b)), which changes from 3,200 tons (2008) to 7,000 tons (2018).

SF<sub>6</sub> possesses superb dielectric strength that is about three times of air at atmospheric pressure (0.1 MPa) and similar to that of transformer oil at 0.3 MPa, which is ascribed to its strong free electrons capture ability, forming heavy ions and preventing the

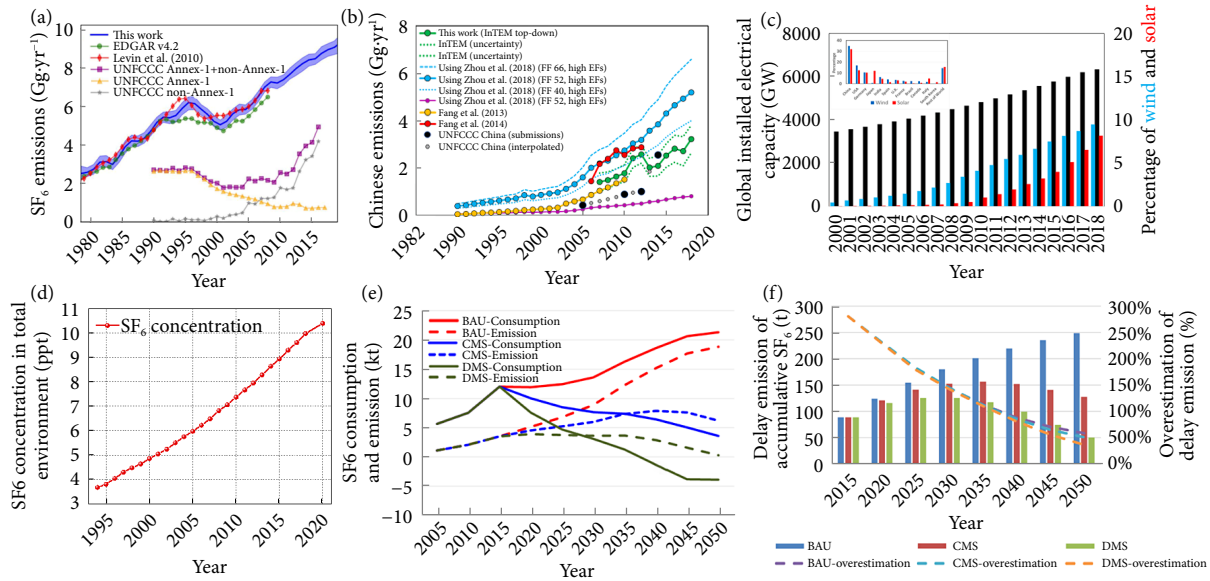
formation of electron flow (Figure 1(c)). Besides, SF<sub>6</sub> has high heat capacity and low viscosity, which is an excellent gas for arc-quenching. Specifically, SF<sub>6</sub> is capable of absorbing arc energy and remaining conductive at low temperatures, which minimizes current chopping and avoids overvoltage<sup>[22–24]</sup>. The arc-quenching capability of SF<sub>6</sub> reaches ~10 times that of air. More importantly, the decomposed particles (SF<sub>x</sub>) could recombine to SF<sub>6</sub> after a high energy arc, making it suitable for repeated current interruption (Figure 1(d))<sup>[22]</sup>. The world's first SF<sub>6</sub> GIS was developed and applied in the United States (US) in 1950s<sup>[25]</sup>. Then the use of SF<sub>6</sub>-based GIE became widespread in the late 1960s in Europe. In 1971, the first 110 kV fully SF<sub>6</sub> insulated GIS and 35 kV circuit breaker (CB) was independently developed in China. Since then, SF<sub>6</sub> plays a dominant role in high voltage, ultrahigh voltage (UHV), and extra-high voltage (EHV) GIE for electricity transmission<sup>[26]</sup>.

### 1.2 SF<sub>6</sub> emission

As introduced in previous section, the emission of SF<sub>6</sub> demonstrates a fast increasing trend, especially in developing countries. According to the report of Simmonds et al.<sup>[27]</sup>, the global emission rate of SF<sub>6</sub> is  $7.3 \pm 0.6$  Gg·yr<sup>-1</sup> (1 Gg=1000 t) in 2008 and it shows an increase of 24% to  $9.04 \pm 0.35$  Gg·yr<sup>-1</sup> in 2018, as shown in Figure 2(a). The maintenance, replacement, and continuous leakage of the GIE are the main reasons for new additional emissions. Accordingly, the SF<sub>6</sub> loading of the atmosphere has increased by a factor of about 15 from 1978 to 2018, achieving a global radiative forcing of  $5.5 \pm 0.1$  mW·m<sup>-2</sup> to the total environment<sup>[28]</sup>. Among all the countries, China shows the largest emission scale with a rapid increase since the beginning of this century (Figure 2(b)). The global installed electrical capacity can be found in Figure 2(c). In 2018, the installed electrical capacity reached 1,899.7 GW, resulting in an estimated emission of SF<sub>6</sub> to be 5.19 Gg, which accounts for about 75.9% of the total SF<sub>6</sub> emission from the electrical aspects<sup>[27]</sup>. It should be noted that China has the largest electrical loading since 2015, corresponding to an annual emission of SF<sub>6</sub> of 3,500 tons



**Fig. 1** The properties and application of SF<sub>6</sub>. (a) The application of SF<sub>6</sub> in various fields. Source: Federal Statistical Office: Survey of particular climate-active materials “sulfur hexafluoride” (SF<sub>6</sub>), Wiesbaden, 2015. (b) The annual SF<sub>6</sub> utilization in China for GIS. Source: Report on alternative technologies of sulfur hexafluoride in the power industry. (c) The dielectric insulation performance of SF<sub>6</sub> compared with oil, solids and air. Reprinted with permission from Ref. [11], © 2021 IEEE. (d,e) The molar fraction of neutral particles versus temperature and time in the SF<sub>6</sub> arc plasma. Reprinted with permission from Ref. [22], © 2016 IOP Publishing Ltd.



**Fig. 2 Emission statistics of SF<sub>6</sub>.** (a) Total emission of SF<sub>6</sub> around the world from 1980 to 2015. Reprinted with permission from Ref. [27], © 2020 Author(s). (b) Estimated emission of SF<sub>6</sub> in China from 1990. Reprinted with permission from Ref. [27], © 2020 Author(s). (c) Global installed electrical capacity (GW). Reprinted with permission from Ref. [27], © 2020 Author(s). (d) Variation of SF<sub>6</sub> concentration in the total environment since 1994. Source: Global Monitoring Laboratory, Earth System Research Laboratories<sup>[7]</sup>. (e) Predictions of SF<sub>6</sub> consumption (solid lines) and emission (dotted lines) (kt) in China under three estimation methods. Reprinted with permission from Ref. [8], © 2018 Author(s). (f) Prediction of the SF<sub>6</sub> delayed emission effect to 2050. Reprinted with permission from Ref. [8], © 2018 Author(s).

this year<sup>[8]</sup>. According to the International Energy Organization, global electricity demand is expected to grow by 30% between 2015 and 2040, and is mainly concentrated in developing countries, which lays a great demand for the development of mature eco-friendly insulation equipment<sup>[8]</sup>.

By contrast, the SF<sub>6</sub> emission in the US and EU countries are relatively reduced in recent decades. As introduced by the US Environmental Protection Agency (USEPA), the SF<sub>6</sub> emission in the USA in 2020 was about 0.236,8 Gg (~5,967.36 Gg CO<sub>2</sub>e)<sup>[29]</sup>. The European Environment Agency has 0.028,6 tons (~720 Gg CO<sub>2</sub>e) SF<sub>6</sub> emission of EU in 2017. The lower emission is attributed to the smaller electrical loadings, more complete SF<sub>6</sub> full-cycle management and already-applied eco-friendly insulation equipment compared to China<sup>[30]</sup>.

The SF<sub>6</sub> atmospheric concentration increased rapidly from ~3.6 ppt (part per trillion) in 1994 to 10.3 ppt in 2020, as shown in Figure 2(d). Moreover, a so-called “delayed emission effect” might happen in China and other countries, leading to a sharp increase in SF<sub>6</sub> emissions (Figure 2(e))<sup>[8]</sup>. It’s attributed to the potential retirement of GIE in the following decades, and the cumulative delayed emission is expected to be 50~249 kt, corresponding to 1.2~6.0 Gt CO<sub>2</sub>e (Figure 2(f)). In this case, the development of eco-friendly insulating gases is also time-critical.

### 1.3 SF<sub>6</sub> reduction policies

SF<sub>6</sub> was labeled as the most potent greenhouse gas in 1995 by the Inter-governmental Panel on Climate Change (IPCC). In 1997, the Kyoto protocol identified SF<sub>6</sub> as the six types of the most greenhouse gases that need to be severely restricted<sup>[31]</sup>. Frankly, the climate concern of SF<sub>6</sub> attributes to its stable molecular structure that endures an extremely atmospheric lifetime. The proficient absorption of infrared radiation further leads to the high GWP of SF<sub>6</sub>. In 2009, the US identified SF<sub>6</sub> as “a threat to the health and welfare of current and future generations due to their effects on world climate” under section 202(a) of the Clean Air Act<sup>[32]</sup>. The

European Union Regulation No. 842-2006 and No. 517/2014 established the rules on the use, recovery and treatment of fluoride gases including SF<sub>6</sub>, which also requires that the emission of SF<sub>6</sub> should be less than 0.1% and the assessment of SF<sub>6</sub> replacements in new MV no later than 2020<sup>[33,34]</sup>. Europe is committed to net zero emission of carbon by 2050. Fluorinated gases used for air conditioning, heat pump, switchgear are participating significantly to carbon emissions. That is why European Commission has decided to propose further improvements to the current F-gases regulation. The draft has been issued in April 2022 and is in discussion for amendments in environmental and industry committees of the European parliament, and country representatives. The vote on the final document by European parliament should happen before the end of 2023. Specifically, the draft proposes to ban gases with GWP > 10 for HV and MV switchgear. However, in order to ensure the continuity of switchgear market, derogation will be given if no solution with gas GWP < 10 is available on the market. In that case, gas with GWP < 2,000 will be accepted. If no solution with GWP gas < 2,000 is available, SF<sub>6</sub> can be accepted. An open tender should be made for switchgear and in case of non-availability of gas GWP < 10, results should be documented for future verification. Table 1 indicates dates of F-gas GWP > 10 prohibitions according to voltage. Besides, the power grid company in Germany, Britain, and Republic of Korea also proposed to utilize SF<sub>6</sub>-free GIE in 2030~2050.

The rapid growth of HV, UHV, EHV projects in China prompts the annual increase of SF<sub>6</sub> usage. In 2012, the GB/T

**Table 1 Prohibition of F-gas GWP > 10 by European Commission.**

Voltage level	Deadline
≤ 24 kV	1 <sup>st</sup> January 2026
≤ 52 kV	1 <sup>st</sup> January 2030
≤ 145 kV, 50 kA	1 <sup>st</sup> January 2028
>145 kV, 50 kA	1 <sup>st</sup> January 2031

28537-2012 entitled the use and handling of SF<sub>6</sub> in high-voltage switchgear and controlgear regulated the treatment during GIE installation, operation, maintenance and end of service<sup>[35]</sup>. Further, the GB/T-32151.2-2015 (Requirements of the greenhouse gas emissions accounting and reporting. Part 2: Power grid enterprise) stipulated the emission accounting of SF<sub>6</sub><sup>[36]</sup>. The State Grid and China Southern Power Grid also formulated relevant SF<sub>6</sub> life-cycle management and control policies, including the establishment of the recycling center, purification and reuse, etc. The SF<sub>6</sub>/N<sub>2</sub> gas mixture based GIE has been put into operation since 2022. The increasingly severe policies and environmental status drive the design and application of eco-friendly insulating gas for next-generation SF<sub>6</sub>-free GIE.

## 2 Development of eco-friendly insulating gas

### 2.1 Basic requirements for eco-friendly gas

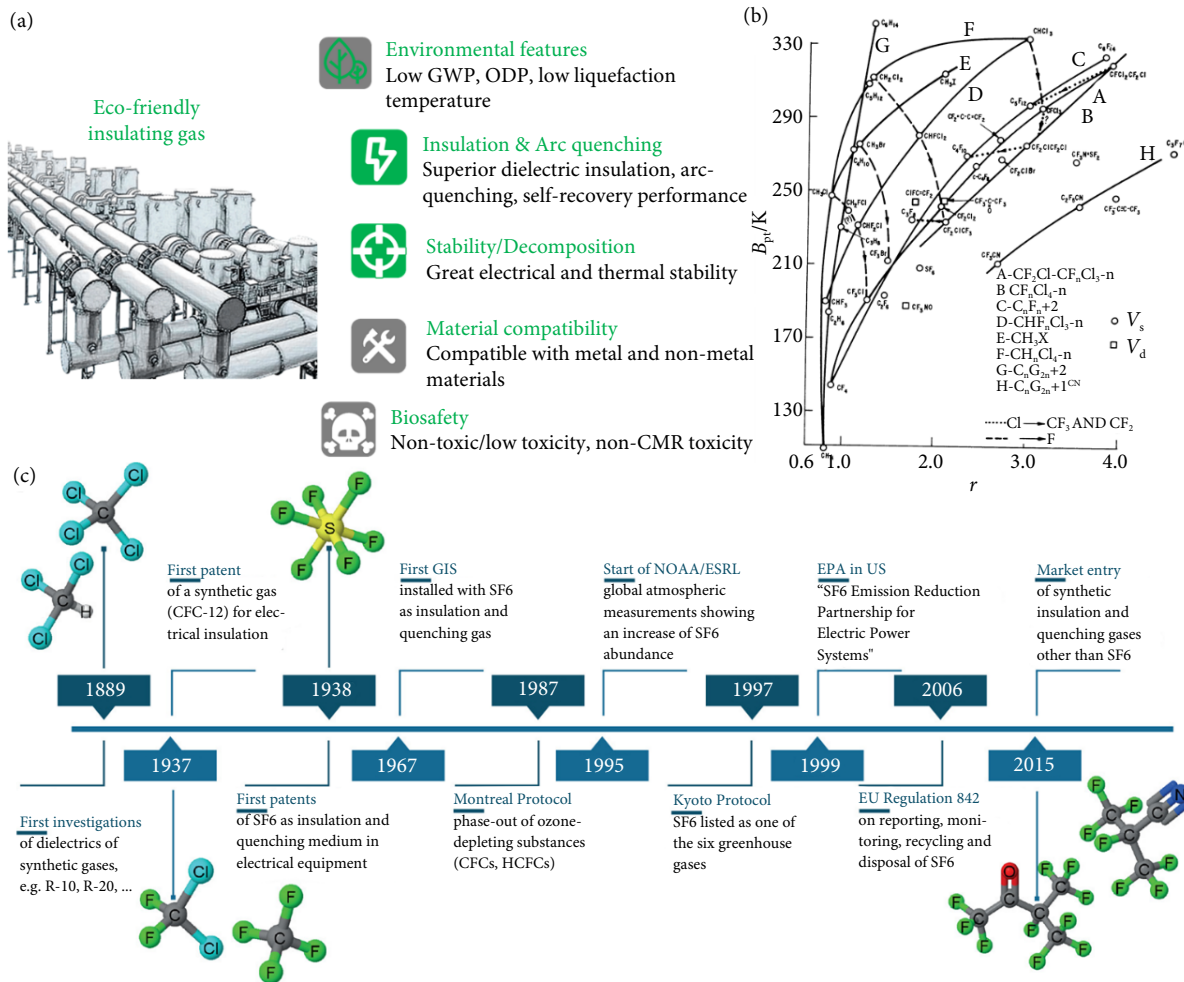
Insulating gas undertakes the mission of electrical insulation and current interruption, which is the core component of GIE. The gas performance essentially determines the operation reliability, service life and maintenance cost of the equipment. The following requirements and selection criteria should be considered for the design or selection of SF<sub>6</sub> substitute gas, as heightened in Figure 3(a).

### Environmental features

The low GWP, no ozone depleting potential (ODP), short atmospheric lifetime and low and are the basic requirements for eco-friendly insulating gas. Specifically, the GWP is related to the absorb ability of infrared rays, the wavelength range of the absorption spectrum and the atmospheric lifetime<sup>[37,38]</sup>. The ODP reflects the potential impact of substance gas escaping into the atmosphere on ozone destruction, which is mainly ascribed to the chlorine, bromine element and nitric oxide<sup>[39,40]</sup>. Besides, low liquefaction temperature is highly desired as it determines the minimum operating temperature of GIE (usually at -15~25°C).

### Insulation & Arc-quenching

High electric strength and superior arc-quenching are the most significant properties of eco-friendly insulating gas. For dielectric insulation, the gas is expected to prevent the formation of electron avalanches or streamers up to high electric field stress<sup>[21]</sup>. The high ionization energy, low electron affinity of the gas molecule is desired to suppress the impact ionization and promotes electron attachment, detachment process that determines the critical electric field. For arc-quenching, the high current interruption capacity, superior heat dissipation and self-recovery performance should be considered. Importantly, the self-recovery ability of gas molecules after arc plasma decomposition determines the repeated quenching property and service life.



**Fig. 3** The basic requirements and classification of eco-friendly insulating gas. (a) The five main properties of advanced insulating gas. (b) The relationship between dielectric strength relative to N<sub>2</sub> and boiling point. Reprinted with permission from Ref. [9], © 1980 IEEE. (c) The timeline regarding substances used for electrical insulation and crucial events concerning the use and development of insulation gases. Reprinted with permission from Ref. [10], © 2018 American Chemical Society.

### Stability/Decomposition

The ideal insulating gas should have excellent thermal, electrical stability. Generally, various early latent defects will inevitably be introduced into the GIE due to transportation, installation, maintenance<sup>[41,42]</sup>. In addition, early insulation faults such as partial discharge (PD) or partial overheating fault (POF) may also occur under long-term operation, which will lead to the decomposition of gas molecules and generate various by-products. The thermal and discharge stability/decomposition condition, by-product type, content as well as the influence of various parameters (gas pressure, mixing ratio, impurities, etc.) should be addressed.

### Material compatibility

Normally, GIE has a relatively long maintenance period (half a year) and decades of service life. More than one hundred different materials are used in GIE, which mainly can be classified into metals and alloys, insulators (epoxy resin), thermoplastics and thermosets, elastomer and adsorbent (desiccant)<sup>[43]</sup>. The high-pressure insulating gas may interact with various materials, inducing gas decomposition and material corrosion owing to the gas-solid thermal/electrical reactions. Investigations on the material's compatibility are necessary to evaluate the stability of eco-friendly gas under long-term or fault conditions. The replacement of incompatible materials or anti-corrosion treatment of solid interface might be conducted.

### Biosafety

Utilization of insulating gas with low toxicity (ideally none) is required for biosafety concerns. The operation and maintenance personnel will inevitably need to contact eco-friendly gas during installation, maintenance, operation. Further, biosafety also determines its application feasibility, waste gas treatment and emission regulations. Besides, the decomposition of insulating gas may generate toxicity by-products, such as the H<sub>2</sub>S, SO<sub>2</sub>, SOF<sub>2</sub>, SO<sub>2</sub>F<sub>2</sub> produced by SF<sub>6</sub>, which also pose potential risks. Thus, evaluation of the safety of eco-friendly insulating gas as well as the decomposition of by-products is necessary.

## 2.2 Classification of eco-friendly gas

In the 1980s, emerging research on the SF<sub>6</sub> substitute gas was initiated by equipment manufacturers such as General Electric (GE). As shown in Figure 3(b), Devins conducted sparking potential measurements for 35 kinds of electron affinity gases, pointing out that the insulation performance of CF<sub>3</sub>—C≡C—CF<sub>3</sub>, C<sub>2</sub>F<sub>5</sub>Cl, C<sub>2</sub>F<sub>5</sub>CN, C<sub>4</sub>F<sub>7</sub>N was superior to SF<sub>6</sub> when applied at 0.2 MPa<sup>[9]</sup>. Later, several novel gases with low GWP have been explored, which can be classified into the following categories.

### Traditional gas

Traditional gas refers to the CO<sub>2</sub>, N<sub>2</sub>, air that commonly exists in daily life. It possesses the advantages such as excellent environmental features, low liquefaction temperature, and excellent biosafety, while its dielectric strength reaches only 30%~38% of SF<sub>6</sub><sup>[44]</sup>. Traditional gas is mainly employed as buffer gas and mixed with electron affinity eco-friendly gas for application currently. Specifically, there might exist synergistic effect between them<sup>[45,46]</sup>. That is, the dielectric performance of gas mixture is higher than the sum of the electric strengths by the mole fractions of its components. The positive synergistic effect is highly desired as the insulation performance of gas mixture will exceed all components. Besides, the utilization of traditional gas at higher working pressure (>0.7 MPa) to achieve better dielectric strength is another strategy for HV SF<sub>6</sub>-free GIE.

### Perfluorocarbons and trifluoroiodomethane

Research on PFCs mainly focuses on CF<sub>4</sub>, C<sub>2</sub>F<sub>6</sub>, C<sub>3</sub>F<sub>8</sub> and *c*-C<sub>4</sub>F<sub>8</sub>, while the first three belong to strong greenhouse gases. *c*-C<sub>4</sub>F<sub>8</sub> has a dielectric strength 1.27 times of SF<sub>6</sub>, the GWP of 8,700 and liquefaction temperature of -6°C<sup>[47,48]</sup>, which needs to be mixed with CO<sub>2</sub>, N<sub>2</sub> or air for application and exhibits little superiority with SF<sub>6</sub>/N<sub>2</sub> gas mixture. Besides, the precipitation of carbon/graphene exists in *c*-C<sub>4</sub>F<sub>8</sub> gas mixture after discharge<sup>[49,50]</sup>. Trifluoroiodomethane (CF<sub>3</sub>I) has similar dielectric strength, lower GWP, atmospheric lifetime compared to *c*-C<sub>4</sub>F<sub>8</sub><sup>[51,52]</sup>. However, the weak C—I bond in its molecular leads to the precipitation of solid iodine in discharges seriously<sup>[53,54]</sup>. In addition, CF<sub>3</sub>I belongs to the carcinogenic, mutagenic, and reprotoxic (CMR) category, bringing potential safety hazards<sup>[55]</sup>. The existing drawbacks stopped the engineering application of PFCs and CF<sub>3</sub>I.

### Fluorinated-nitrile, ketones

In 2015, Minnesota Mining and Manufacturing (3M) presented fluorinated nitrile (C<sub>4</sub>F<sub>7</sub>N) and fluorinated ketones (C<sub>5</sub>F<sub>10</sub>O and C<sub>6</sub>F<sub>12</sub>O) as the latest eco-friendly insulating gas<sup>[56,57]</sup>. Specifically, C<sub>4</sub>F<sub>7</sub>N has a dielectric strength of 2.2 times that of SF<sub>6</sub>, a GWP of 2090, and an atmospheric lifetime of years. The ketones (C<sub>5</sub>F<sub>10</sub>O and C<sub>6</sub>F<sub>12</sub>O) also demonstrate superior environmental features (GWP of 1) and dielectric strength (1.4 and > 2 times of SF<sub>6</sub>). Owing to the relatively high liquefaction temperature, fluorinated nitrile and ketones have been introduced as admixtures to traditional gas for engineering application. The CO<sub>2</sub>, technical air, O<sub>2</sub> is mainly utilized to obtain the binary or ternary gas mixture. For example, C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> with 6%, 10% C<sub>4</sub>F<sub>7</sub>N fulfills the minimum operating temperature of -25°C, -10°C (0.6 MPa), and the GWP value of gas mixture decreased to 462 and 690<sup>[56]</sup>. The fluorinated ketone based gas mixture is mainly used for MV GIE with 7%~14% C<sub>5</sub>F<sub>10</sub>O or 3%~6% C<sub>6</sub>F<sub>12</sub>O added<sup>[58,59]</sup>. For HV application, the mole fraction of ketones and the minimum operation temperature is further limited.

### Hydrofluoro-Olefins


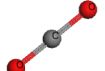
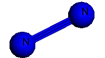
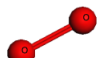
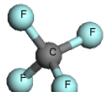
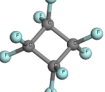
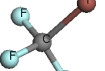
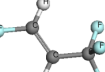
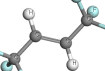
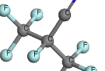
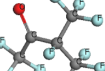
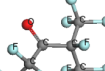
Hydrofluoro-Olefins (HFOs) are mainly used as refrigerants to replace the PFCs or saturated hydrofluorocarbons (HFCs) at present, in which HFO-1234ze(E), HFO-1336mzz(E) is proposed as eco-friendly insulating gas recently<sup>[60-62]</sup>. Specifically, HFO-1234ze(E) and HFO-1336mzz(E) have a GWP of 1 and 18, a dielectric strength of 0.81~0.98 and 1.63~1.86 times of SF<sub>6</sub>, and a liquefaction temperature of -19.2°C and 7.58°C, respectively<sup>[63,64]</sup>. Thus, HFO-1234ze(E) can be used as pure gas in MV GIE down to -15°C. Besides, the acute toxicity of HFO-1234ze(E) has been systematically evaluated. Therefore, HFOs appear promising application potential for MV GIE that is mainly located in urban distribution networks, rail transit, buildings and other people crowded places.

Table 2 and Figure 3(c) summarized the basic parameters and development paths of eco-friendly insulating gas<sup>[10]</sup>. To date, it is still challenging to screen one gas with comprehensive performance as SF<sub>6</sub>. There exists a trade-off or conflict between the environmental features and stability, dielectric strength and liquefaction temperature, etc.

## 2.3 Molecular design of eco-friendly gas

The key to designing new insulating gas is to predict its macro performance through the parameters at the molecular level. The general framework is shown in Figure 4. First, the possible gas molecular structure is preset, and then its performance is calculated

Table 2 The basic parameters of common eco-friendly insulating gas.

Chemical formula	Molecular structure	GWP (100 years)	Atmospheric lifetime (year)	Liquefaction temperature(°C)	Dielectric strength relative to SF <sub>6</sub> (E/N) <sub>crit</sub>	LC50 (rat, 4 h, μL/L)
SF <sub>6</sub>		25,200	3,200	-64	1	—
CO <sub>2</sub>		1	—	-78.5	0.35	—
N <sub>2</sub>		—	—	-196	0.38	—
O <sub>2</sub>		—	—	-183	0.33	—
CF <sub>4</sub>		6,630	50,000	-128	0.41	—
c-C <sub>4</sub> F <sub>8</sub>		8,700	3,200	-6	1.27	—
CF <sub>3</sub> I		0.4	0.005	-21.8	1.2	160,000
HFO-1234ze(E)		<1	0.045	-19.2	0.81~0.98	>207,000
HFO-1336mzz(E)		18	0.06	7.58	1.63~1.86	25,400~49,000
C <sub>4</sub> F <sub>7</sub> N		2,090	22	-4.7	2.2	12,500~15,000
C <sub>5</sub> F <sub>10</sub> O		<1	0.044	26.9	1.4	20,000
C <sub>6</sub> F <sub>12</sub> O		1	0.014	49	2.2	>100,000

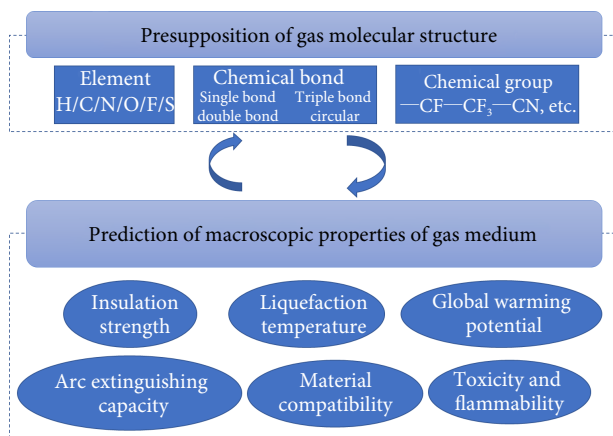


Fig. 4 General framework for molecular design of new insulating gases.

through an appropriate model. If the performance does not meet the requirements, the molecular structure is adjusted, and the gas molecules meeting the performance are obtained iteratively. In the process of molecular design, it is quite difficult to establish a performance prediction model due to missing data references. At present, the main prediction is for insulation strength or liquefaction temperature.

#### Performance prediction model

The first problem of the prediction model is how to select appropriate predictors. Most of the models reported at present directly give the predictors. Common insulation prediction factors are as follows:

- (1) Ground state electric dipole momentum, average electric polarizability, and electronic affinity energy (EA)<sup>[65]</sup>.
- (2) Four predictors for polar molecules are electric dipole

moment, electron number, average static electronic polarizability, and adiabatic ionization energy. Four predictors for nonpolar molecules are average static electronic polarizability, adiabatic ionization energy, adiabatic electron affinity, and vertical electron affinity<sup>[66]</sup>.

(3) Integral of the optical absorption (IOA)<sup>[67]</sup>.

(4) The total surface area of the molecule, statistical variance, degree of balance, average deviation, molecular polarizability, absolute electronegativity, and hardness<sup>[68-71]</sup>.

(5) Molecular diameter (van der Waals surface), 1<sup>st</sup> ionization energy, molecular electronegativity<sup>[72]</sup>.

(6) Ionization energy, polarizability<sup>[73]</sup>.

(7) General interaction properties function (GIPF) parameter of isosurface with electron localization function (ELF) of 0.2<sup>[74]</sup>.

(8) Molecular volume, polar surface area, surface positive average potential, surface negative average potential, molecular polarity index<sup>[75]</sup>.

(9) Molecular surface electrostatic potential, molecular volume, molecular surface area, polarizability<sup>[76]</sup>.

(10) Electric dipole moment, positive surface area, average static electronic polarizability, molecular volume, highest occupied molecular orbital (HOMO), lowest unoccupied molecular orbital (LUMO), molecular mass<sup>[77]</sup>.

Using these predictors and the reported insulation strength data, usually relative to the insulation strength of SF<sub>6</sub> gas, the corresponding relationship between the predictors and gas insulation strength can be established by regression analysis. In addition, these predictors can be obtained through density functional theory (DFT) calculation. In this way, the insulation strength of unknown gas can be predicted.

Characteristics of new gas

Through molecular design, gases that meet the requirements in theory can be obtained, and the performance needs to be determined through experiments. However, most of the designed gases

cannot be bought and research on these gases can only depend on artificial synthesis. One typical example is the CF<sub>3</sub>SO<sub>2</sub>F proposed by Zhou et al., which has advantages in insulation, liquefaction through a prediction model and it was successfully synthesized in the laboratory<sup>[69,78]</sup>. Further, the electron swarm parameters of CF<sub>3</sub>SO<sub>2</sub>F gas were obtained by the steady-state Townsend (SST) experiment<sup>[79]</sup>, and its dielectric strength under higher pressure was obtained through AC breakdown experiment<sup>[80]</sup>. Recently, the toxicity study of this gas has also reported preliminary results, indicating that CF<sub>3</sub>SO<sub>2</sub>F gas will be listed as a toxic compound by most standards<sup>[81]</sup>. This poses challenges for its application as an eco-friendly gas. It also shows that molecular design needs to consider multi-dimensional features, not only insulation or liquefaction.

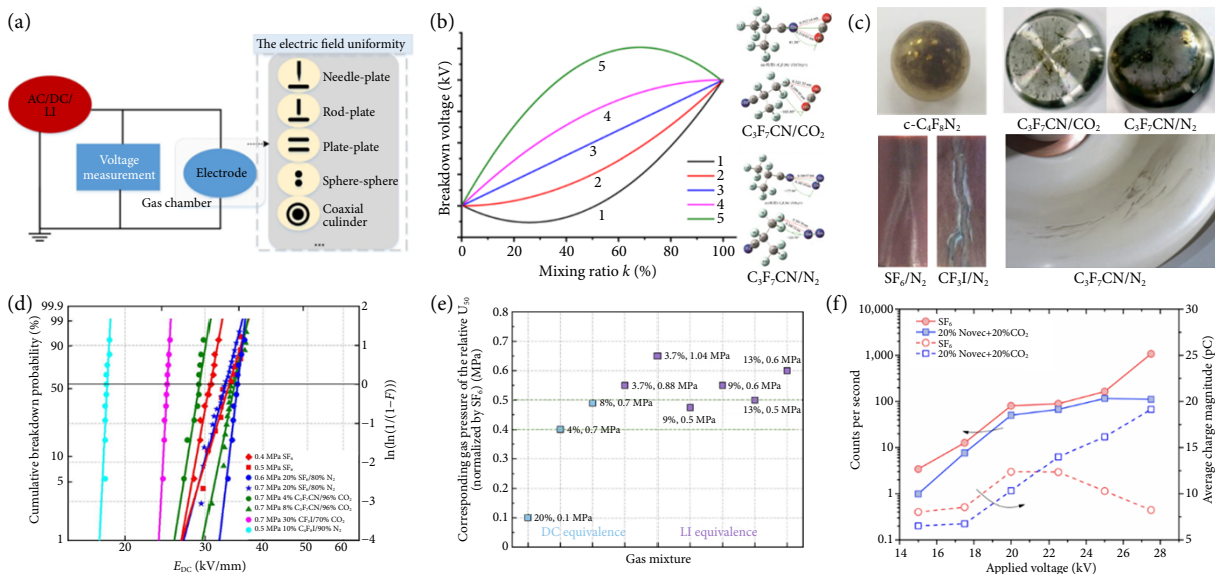
3 Assessment of eco-friendly insulating gas

3.1 Dielectric properties

Dielectric properties are the foundation of eco-friendly insulating gas. Research on the insulation performance of eco-friendly insulating gas mainly focuses on its breakdown, PD, and surface flashover characteristics. The GIL or GIS has a coaxial structure that is dominated by a quasi-uniform electric field. Defects like protrusions or metallic particles also exist. Under different voltage forms and various electric fields, eco-friendly gases show different performance. Figure 5(a) depicts the methods and typical electrode system for the breakdown voltage measurement<sup>[82]</sup>.

Alternating current (AC) breakdown

The AC breakdown voltage of PFCs with better insulating properties and low GWP values such as *c*-C<sub>4</sub>F<sub>8</sub>, C<sub>3</sub>F<sub>8</sub>, C<sub>2</sub>F<sub>6</sub> have been tested<sup>[83]</sup>. Buffer gas such as N<sub>2</sub> or CO<sub>2</sub> is employed owing to the high boiling point. The synergistic effect between fluorocarbon main insulating and buffer gas should be considered, as it offers



**Fig. 5 The dielectric properties of eco-friendly insulating gases.** (a) Diagram of the breakdown voltage measurement. Reprinted with permission from Ref. [83], © 2020 Author(s). (b) Synergy relationship of gas mixture: negative synergistic effect  $C > 1$  (curves 1 and 2), linear relationship  $C = 1$  (curve 3), and positive synergistic effect  $C < 1$  (curves 4 and 5). Reprinted with permission from Refs. [83, 110], © 2019 Author(s). (c) The soot layer and trace on electrodes or insulators after gas discharge. Reprinted with permission from Refs. [19, 114], © 2018, IEEE. (d) The Weibull distribution of DC dielectric strength of various eco-friendly insulating gases. Reprinted with permission from Ref. [20], © 2021, IEEE. (e) Equivalence of breakdown voltages between C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> and SF<sub>6</sub> at DC and LI voltage. Reprinted with permission from Ref. [19], © 2020 Author(s). (f) The PD number and PD magnitude for the 20% C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> and SF<sub>6</sub> under different voltages. Reprinted with permission from Ref. [101], © 2018 IEEE.

additional insulation performance (Figure 5(b)). The insulation performance of the *c*-C<sub>4</sub>F<sub>8</sub> gas under uniform electric field is 1.18~1.25 times than that of SF<sub>6</sub>, and synergistic effect was found when PFCs combined with CO<sub>2</sub>, N<sub>2</sub>, CF<sub>4</sub>, respectively<sup>[84–87]</sup>. The dielectric strength of gas mixture with 60% CF<sub>3</sub>I can reach that of SF<sub>6</sub><sup>[88–90]</sup>, and CO<sub>2</sub> possesses better synergistic effect than that of N<sub>2</sub><sup>[91,92]</sup>. However, the obvious solid by-product is generated (mainly includes I and C) under discharge (Figure 5(c)), which is very detrimental to gas self-recovery and mainly limits its application as the oxidation of materials due to the presence of iodine could not be ignored<sup>[53,54]</sup>.

In 2015, Asea Brown Boveri (ABB) proposed the utilization of C<sub>5</sub>F<sub>10</sub>O or C<sub>6</sub>F<sub>12</sub>O in combination with technical air as insulating medium<sup>[93]</sup>. The breakdown voltage of the gas mixture containing 5.2% C<sub>5</sub>F<sub>10</sub>O at 0.7 MPa can reach 95%, 80% of SF<sub>6</sub> at 0.45 MPa, 0.6 MPa. The addition of 3% C<sub>6</sub>F<sub>12</sub>O to N<sub>2</sub> can bring double breakdown voltage of pure N<sub>2</sub><sup>[94,95]</sup>. It is considered that fluorinated ketone is more suitable for MV GIE owing to the relatively high liquefaction temperature. In 2016, GE reported the AC breakdown characteristics of C<sub>4</sub>F<sub>7</sub>N gas mixture for the first time<sup>[96]</sup>. The gas mixture with 20% C<sub>4</sub>F<sub>7</sub>N has similar dielectric strength to that of SF<sub>6</sub>, and CO<sub>2</sub> or air demonstrates a better synergistic effect compared to that of N<sub>2</sub>. The breakdown voltage of 3.7% C<sub>4</sub>F<sub>7</sub>N/96.3% CO<sub>2</sub> at 0.64 MPa, 0.76 MPa can reach the level of SF<sub>6</sub> at 0.55 MPa and 0.65 MPa<sup>[97–99]</sup>.

HFOs such as HFO-1234ze(E), HFO-1336mzz(E) are also focused. The former has comparable insulation performance to SF<sub>6</sub> at 130 kPa<sup>[100]</sup>. The positive synergistic effect between HFO-1234ze(E) and SF<sub>6</sub> was found, and the gas mixture with 40%~60% HFO-1234ze(E) demonstrates 1.2 times dielectric strength of SF<sub>6</sub><sup>[101]</sup>. The AC breakdown voltage of HFO-1336mzz(E)/CO<sub>2</sub> with 25~30% HFO-1336mzz(E) can reach 75%~85% of SF<sub>6</sub> under the quasi-uniform electric field, and the relative dielectric strength can be significantly improved by increasing mixing ratio. There exists a favorable synergistic effect between HFO-1336mzz(E) and CO<sub>2</sub>, and the breakdown voltage of HFO-1336mzz(E)/CO<sub>2</sub> mixture under the highly non-uniform electric field demonstrates a “hump” phenomenon with the increase of gas pressure, especially at 0.2 MPa<sup>[62]</sup>.

#### Direct current (DC) Breakdown

The characteristics of gas under DC electric field are different from that of AC. The insulation performance of 20% CF<sub>3</sub>I/80% N<sub>2</sub> and 30% CF<sub>3</sub>I/70% N<sub>2</sub> at 0.15–0.25 MPa did not increase remarkably with gas pressure<sup>[20]</sup>. The negative DC dielectric strength of 4% C<sub>4</sub>F<sub>7</sub>N/96% CO<sub>2</sub>, 8% C<sub>4</sub>F<sub>7</sub>N/92% CO<sub>2</sub> at 0.7 MPa reached 81.21% and 96.5% of SF<sub>6</sub> at 0.5 MPa, respectively<sup>[102,103]</sup>. Moreover, the DC insulation properties were found to be sensitive to the electrode surface roughness<sup>[104–106]</sup>. The DC breakdown voltage changed to saturation with the increase of gas pressure, especially for electrodes with larger surface roughness. The 6% C<sub>4</sub>F<sub>7</sub>N/94% CO<sub>2</sub> mixture was less sensitive to the electrode surface roughness than SF<sub>6</sub> and 20% SF<sub>6</sub>/80% N<sub>2</sub>(Figure 5(d))<sup>[106]</sup>. In addition, HFO-1336mzz(E)/CO<sub>2</sub> has a higher negative DC dielectric strength than HFO-1336mzz(E)/N<sub>2</sub>, which can occupy 93.03% and 86.39% of 20%SF<sub>6</sub>/80%N<sub>2</sub> at 0.7 MPa<sup>[107]</sup>.

Defects like protrusions or metallic particles will lead to strong local non-uniform electric fields, resulting in a decrease in dielectric strength. It was reported that the DC breakdown characteristics of C<sub>4</sub>F<sub>7</sub>N/N<sub>2</sub> were strongly affected by polarity<sup>[106,108,109]</sup>. At the same gas pressure, the negative DC breakdown voltage was much higher than the positive, and their difference increased with pres-

sure. The magnitude of the negative breakdown voltage increased with pressure, whereas the positive breakdown voltage displayed the opposite behavior.

#### Lightning impulse (LI) breakdown

The insulation characteristics of fluorocarbon gas also possess a polarity effect under lightning impulse breakdown. For example, the negative LI breakdown voltage of *c*-C<sub>4</sub>F<sub>8</sub>/N<sub>2</sub> was lower than that of positive. This was ascribed to the difference in free electrons' speed and kinetic energy. The free electrons in a negative lightning strike had a higher velocity, making the gas molecules hard to capture. The synergistic effect of C<sub>3</sub>F<sub>8</sub>/N<sub>2</sub>, C<sub>2</sub>F<sub>6</sub>/N<sub>2</sub> was found to be inferior to that of SF<sub>6</sub>/N<sub>2</sub>, so as to the CF<sub>3</sub>I gas mixture<sup>[110]</sup>. The positive LI breakdown voltage of 5% C<sub>5</sub>F<sub>10</sub>O/95% air at 0.7 MPa reached that of pure SF<sub>6</sub> at 0.4 MPa<sup>[93,94]</sup>. For C<sub>4</sub>F<sub>7</sub>N, the LI insulation performance of 3.7% C<sub>4</sub>F<sub>7</sub>N/96.3% CO<sub>2</sub> at 0.88 MPa was equal to that of SF<sub>6</sub> at 0.55 MPa, and higher increase of pressure at 1.04 MPa brought little enhancement (reached SF<sub>6</sub> at 0.65 MPa)<sup>[97,111]</sup>. The equivalence of breakdown voltage between C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> and SF<sub>6</sub> gas at DC and LI voltage was summarized in Figure 5(e)<sup>[19]</sup>.

#### Partial discharge

Fault-induced PD inside the GIE will cause electromagnetic radiation, noise, gas decomposition and materials deterioration. The gas insulating medium with superior PD withstand performance is highly desired. For PFCs, the partial discharge inception voltage (PDIV) of *c*-C<sub>4</sub>F<sub>8</sub>/N<sub>2</sub> with 15%~20% *c*-C<sub>4</sub>F<sub>8</sub> reaches 65%~70% that of SF<sub>6</sub><sup>[112]</sup>. The PDIV of CF<sub>3</sub>I/CO<sub>2</sub> was higher than that of CF<sub>3</sub>I/N<sub>2</sub>, and the 30% CF<sub>3</sub>I/70% CO<sub>2</sub> was about 70% of SF<sub>6</sub> at 0.1 MPa<sup>[113]</sup>. The 20% C<sub>4</sub>F<sub>7</sub>N/80% CO<sub>2</sub> shows slightly higher PDIV with respect to SF<sub>6</sub>, and SF<sub>6</sub> has much more PDs with low magnitude while C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> demonstrated fewer PDs with higher magnitude (Figure 5(f))<sup>[98,99]</sup>. The PDIV of C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> under an extremely non-uniform electric field demonstrated “hump effect”, and the inhibition ability on negative PD is better than that of SF<sub>6</sub>/N<sub>2</sub>. For DC voltage, the negative polarity PDIV of C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> is higher than that of positive polarity, which is greatly affected by the non-uniformity coefficient of the electric field<sup>[114]</sup>. In addition, the C<sub>5</sub>F<sub>10</sub>O/air possessed higher PD amplitude under positive polarity, while for SF<sub>6</sub> it is under negative polarity.

Overall, fluorocarbon gas is proven to be sensitive to electric field inhomogeneity. The PD discharge repetition rate and average discharge magnitude of eco-friendly insulating gas with equivalent insulation performance to SF<sub>6</sub> are higher. The inhibition ability of negative PD is better than that of positive ones. In addition, the relative PDIV of fluorocarbon gas under high pressure is lower than that of low-pressure conditions. Considering that the proportion of buffer gases such as CO<sub>2</sub> with weak insulation performance is high, the PD characteristics are similar to CO<sub>2</sub> to some extent. In terms of engineering application, it is necessary to avoid the introduction of extremely uneven electric fields, and to strengthen PD monitoring and analysis in operation and maintenance.

#### Flashover

Surface flashover properties that are important and complex for gas-solid insulation design have also been evaluated. For PFCs, it was found that the flashover voltage of *c*-C<sub>4</sub>F<sub>8</sub> was 1.2 times that of SF<sub>6</sub> with obvious polarity. The AC flashover voltage of CF<sub>3</sub>I/N<sub>2</sub> is higher than that of SF<sub>6</sub>/N<sub>2</sub>, while it was lower under DC stress. For C<sub>4</sub>F<sub>7</sub>N, the AC surface flashover voltage of 9% C<sub>4</sub>F<sub>7</sub>N/91% CO<sub>2</sub>



under 0.6 MPa could reach that of SF<sub>6</sub> at 0.5 MPa<sup>[115,116]</sup>. The negative DC flashover voltage of 4% C<sub>4</sub>F<sub>7</sub>N/96% CO<sub>2</sub>, 8% C<sub>4</sub>F<sub>7</sub>N/92% CO<sub>2</sub> at 0.7 MPa were 96.06% and 101.50% of SF<sub>6</sub> at 0.5 MPa. In addition, the LI surface flashover voltage shows a saturated growth trend with pressure and mixing ratio, and the gas mixture with 5%, 9%, and 13% C<sub>4</sub>F<sub>7</sub>N can reach 70%, 80%, 90% of SF<sub>6</sub> under the same conditions. The surface discharge pattern in C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> and SF<sub>6</sub> has no obvious difference<sup>[116]</sup>. It should be noted that the accumulation of surface charge under DC conditions might cause electric field distortion, bringing specific surface flashover at lower voltage<sup>[117]</sup>. Relevant tests also found that the pot insulator flashover mostly occurred at the concave side, and the flashover voltage of 9% C<sub>4</sub>F<sub>7</sub>N/91% CO<sub>2</sub> at 0.6 MPa reached that of SF<sub>6</sub> at 0.5 MPa.

On the whole, current research has confirmed the superior insulation withstands characteristics of eco-friendly gases. Table 3 further summarized and compared the dielectric properties of various eco-friendly insulating gases. Relevant data provide important references for the design and optimization of GIE. For HV application scenario, the increase of gas pressure or minimum operating temperature (an increase of fluorocarbon gas content) is needed owing to their inferior dielectric strength. Meanwhile, the introduction of an inhomogeneous electric field should be avoided considering the reduction of relative insulation performance at high pressure. Further explorations on the synergistic effect mechanism should be addressed.

### 3.2 Arc-quenching

One of the essential elements to consider when applying eco-friendly gases in next-generation SF<sub>6</sub>-free equipment is their arc-quenching ability. A good arc-quenching medium must present not only high thermal conductivity to cool the arc quickly but also high dielectric strength to facilitate the transient recovery voltage and to avoid the reignition of the arc<sup>[135]</sup>. To evaluate the arc-quenching performance of eco-friendly gases, many previous works have been done through numerical and experimental approaches.

#### Particle compositions

High-temperature arc is a kind of thermal plasma that has different particle compositions at different temperatures and pressures. The composition of an arc also plays an essential role in the calculation of arc plasma properties, such as thermodynamic properties and transport coefficients<sup>[136]</sup>. The computational results indicate that many eco-friendly gases, such as CF<sub>3</sub>I<sup>[137]</sup>, C<sub>4</sub>F<sub>8</sub><sup>[138]</sup>, C<sub>4</sub>F<sub>7</sub>N<sup>[139–141]</sup>, and C<sub>5</sub>F<sub>10</sub>O<sup>[139,142–144]</sup>, cannot recombine into itself after dissociation in arc plasmas. Most composition calculations assume that arc plasmas are at local thermodynamic equilibrium (LTE) and all species are gaseous. However, this is not always valid for eco-friendly gases. For instance, C<sub>4</sub>F<sub>7</sub>N and C<sub>5</sub>F<sub>10</sub>O can produce condensed species, i.e., graphite in arc plasmas (Figure 6(a)), and the condensation temperature of graphite decreases more or less with the increase of buffer gases<sup>[142,145]</sup>. To yield graphite-free products at room temperature, O<sub>2</sub> can be mixed with C<sub>4</sub>F<sub>7</sub>N and C<sub>5</sub>F<sub>10</sub>O mixtures to completely constrain the production of graphite<sup>[139]</sup>. Since the condensed species exist only at low temperatures, it is necessary to consider the departure from LTE, which requires the calculation of non-LTE two-temperature (2T) plasma compositions<sup>[146,147]</sup>. It is found that the condensation temperatures of condensed species in eco-friendly gaseous arcs do not vary monotonously with the non-equilibrium degree in arc plasmas, and the balance effect can be observed between electron temperature  $T_e$  and heavy particle temperature  $T_h$  at low value of non-equilibrium degree<sup>[139]</sup>.

#### Thermodynamic properties

Based on the particle compositions, the thermodynamic properties including mass density, specific heat, enthalpy, entropy, etc., can be calculated directly according to their thermodynamic definitions<sup>[147]</sup>. Generally, a gas with high specific heat has good ability to cool the hot arc during the arc extinction. In comparison to SF<sub>6</sub>, C<sub>5</sub>F<sub>10</sub>O and C<sub>5</sub>F<sub>10</sub>O-CO<sub>2</sub> mixtures present higher specific heat except in the medium temperature range<sup>[142]</sup>. Some previous works reveal that the thermal interruption performance of a gas can be evaluated by the temperature dependence of the product of specific heat and mass density (i.e.,  $\rho C_p$ )<sup>[137,148]</sup>. C<sub>4</sub>F<sub>8</sub>-CO<sub>2</sub> arc shows higher peaks of  $\rho C_p$  at high temperatures and lower peaks at low temperatures than SF<sub>6</sub><sup>[138]</sup>. C<sub>5</sub>F<sub>10</sub>O arc presents a similar characteristic<sup>[143]</sup>. It is also found that the mixing of buffer gases (e.g., N<sub>2</sub> and CO<sub>2</sub>) can gradually erase the peaks of  $\rho C_p$  in the C<sub>4</sub>F<sub>7</sub>N arc (Figure 6(b))<sup>[141]</sup>, which inevitably affects the arc cooling rate during the arc-quenching process. Most previous works on the thermodynamic properties of eco-friendly gases only consider gaseous species. However, the production of graphite in C<sub>5</sub>F<sub>10</sub>O arc has an influence on the mass density and specific enthalpy in the low-temperature range<sup>[142]</sup>.

#### Transport coefficients

Arc plasma modeling requires not only thermodynamic properties but also transport coefficients. The transport coefficients including electrical conductivity, viscosity, and thermal conductivity can be determined by the collision integrals based on the Chapman-Enskog method<sup>[149]</sup>. The condensed species in the arc plasmas of eco-friendly gases, e.g., graphite, are usually neglected in calculating collision integrals and transport coefficients. Since eco-friendly gases are large molecules that have rich products in arc plasmas, many complex chemical reactions have to be considered in the calculation of thermal conductivity. These reactions especially the dissociation and ionization reactions lead to peaks of thermal conductivity<sup>[138,140,143,144]</sup>. The arc cooling rate strongly depends on the peaks of thermal conductivity, particularly on the peak at the highest temperature. The arc decays fast above this highest temperature and slowly below this temperature<sup>[150]</sup>. The mixing of buffer gases, such as CO<sub>2</sub>, N<sub>2</sub>, and O<sub>2</sub>, also has an effect on the transport coefficients of eco-friendly gaseous arcs (Figure 6(c)). For example, the mixing of CO<sub>2</sub> or N<sub>2</sub> shows a significant effect on the viscosity of C<sub>4</sub>F<sub>8</sub>-CO<sub>2</sub>, C<sub>4</sub>F<sub>7</sub>N-CO<sub>2</sub>, and C<sub>4</sub>F<sub>7</sub>N-N<sub>2</sub> arcs<sup>[138,140]</sup>. However, the buffer gases do not show a distinct influence on the electrical conductivity of C<sub>4</sub>F<sub>7</sub>N arcs if only small proportion of buffer gas is mixed<sup>[140]</sup>.

#### Radiation coefficients

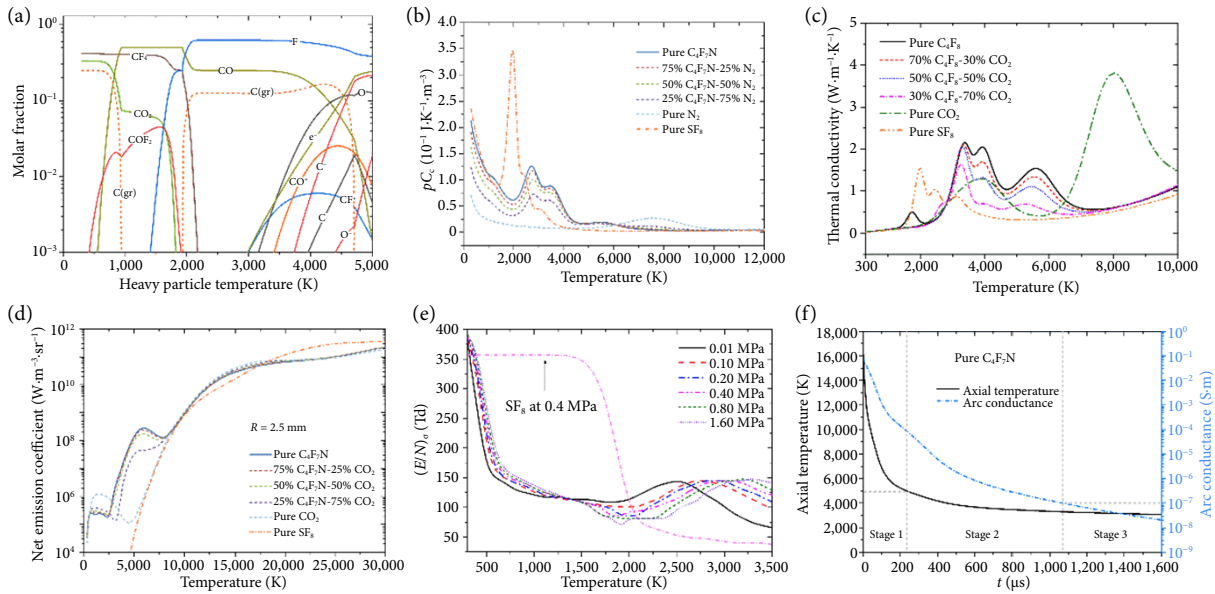
Radiation plays an import role in arc extinction. In high-voltage circuit breakers (HVCBs), the energy loss due to radiation increases the cooling rate of the arc during the current zero and is responsible for the ablation of the nozzle<sup>[138]</sup>. Therefore, the radiation properties of an arc are required to understand the arc decaying process<sup>[140]</sup>. However, solving radiation transport equations is rather complicated and time-consuming, as the radiation transfer strongly depends on the computational geometry and gas types, and is a function of wavelength spanning from the infrared to the far ultraviolet region of the spectrum<sup>[138]</sup>. As a result, a simplified model, namely net emission model, is usually used in arc modeling. The corresponding coefficients in this model are net emission coefficients (NEC) which can be calculated by considering all the atomic and molecular radiations in arc plasmas<sup>[151–154]</sup>. It is found that the NEC of C<sub>4</sub>F<sub>8</sub>-CO<sub>2</sub> arcs is slightly larger than that of SF<sub>6</sub><sup>[138]</sup>.

Table 3 The dielectric properties of eco-friendly insulating gas under different voltage types.

Gas	Voltage type	Dielectric properties
c-C <sub>4</sub> F <sub>8</sub>	AC	◆ Sphere-plate electrode, 0.2–0.5 MPa, 50% c-C <sub>4</sub> F <sub>8</sub> /50% N <sub>2</sub> , 90% that of SF <sub>6</sub> <sup>[118]</sup> .
		◆ Sphere-plate electrode, 0.1 MPa, 20% c-C <sub>4</sub> F <sub>8</sub> /80% N <sub>2</sub> or CO <sub>2</sub> , 67%(N <sub>2</sub> ), 68%(CO <sub>2</sub> ) that of SF <sub>6</sub> ; 30% c-C <sub>4</sub> F <sub>8</sub> /70% N <sub>2</sub> or CO <sub>2</sub> , 0.1 MPa, 75%(N <sub>2</sub> ), 79%(CO <sub>2</sub> ) that of SF <sub>6</sub> <sup>[119]</sup> .
	LI	◆ Needle-plate electrode, 0.1–0.3 MPa, 20% c-C <sub>4</sub> F <sub>8</sub> /80% N <sub>2</sub> , 60%–90% that of 20%SF <sub>6</sub> /N <sub>2</sub> <sup>[120]</sup> .
	DC	◆ Sphere-plate electrode, 0.1 MPa, 10% c-C <sub>4</sub> F <sub>8</sub> /90% N <sub>2</sub> or CO <sub>2</sub> , positive LI breakdown voltage higher than that of negative, 60% that of SF <sub>6</sub> <sup>[121]</sup> .
Surface flashover	◆ Sphere-plate electrode, 0.1–0.5 MPa, 20% CF <sub>3</sub> I/80% N <sub>2</sub> , 62% that of SF <sub>6</sub> ; 10% CF <sub>3</sub> I/90% N <sub>2</sub> , 43% that of SF <sub>6</sub> <sup>[122]</sup> .	
	◆ AC, 0.1 MPa, 30% c-C <sub>4</sub> F <sub>8</sub> /70% N <sub>2</sub> , 73% that of SF <sub>6</sub> <sup>[123]</sup> .	
CF <sub>3</sub> I	AC	◆ DC, 0.1–0.5 MPa, 20% CF <sub>3</sub> I/80% N <sub>2</sub> , 71% that of SF <sub>6</sub> ; 10% CF <sub>3</sub> I/90% N <sub>2</sub> , 65% that of SF <sub>6</sub> <sup>[122]</sup> .
		◆ Sphere-plate electrode, 0.1–0.3 MPa, 30% CF <sub>3</sub> I/70% N <sub>2</sub> , 92%–98% that of SF <sub>6</sub> <sup>[124]</sup> .
	LI	◆ Needle-plate, 0.3 MPa, 30% CF <sub>3</sub> I/70% N <sub>2</sub> , 85%–90% that of SF <sub>6</sub> <sup>[88]</sup> .
	DC	◆ Needle-plate electrode, 0.1–0.3 MPa, 30% CF <sub>3</sub> I/70% N <sub>2</sub> , lower than 20% SF <sub>6</sub> /80% N <sub>2</sub> (80%–86%) <sup>[110]</sup> .
Surface flashover	◆ Plate-plate electrode, 0.1–0.2 MPa, 30% CF <sub>3</sub> I/70% N <sub>2</sub> , similar to that of 20% SF <sub>6</sub> /80% N <sub>2</sub> <sup>[91]</sup> .	
	◆ LI, 0.1–0.3 MPa, 30% CF <sub>3</sub> I/70% N <sub>2</sub> , similar to 20% SF <sub>6</sub> /80% N <sub>2</sub> under negative LI, higher than 20% SF <sub>6</sub> /80% N <sub>2</sub> under positive LI <sup>[110]</sup> .	
C <sub>4</sub> F <sub>7</sub> N	AC	◆ Sphere-plate electrode, 0.1–0.7 MPa, 13% C <sub>4</sub> F <sub>7</sub> N/87% CO <sub>2</sub> , >90% that of SF <sub>6</sub> ; 9% C <sub>4</sub> F <sub>7</sub> N/91% CO <sub>2</sub> , >80% that of SF <sub>6</sub> <sup>[125]</sup> .
		◆ Sphere-sphere electrode, 0.1–0.3 MPa, 8% C <sub>4</sub> F <sub>7</sub> N/92% CO <sub>2</sub> , 72.8%(0.3 MPa)–76.5%(0.1 MPa) that of SF <sub>6</sub> <sup>[99]</sup> .
	LI	◆ Needle-plate electrode, 0.1–0.3 MPa, 8% C <sub>4</sub> F <sub>7</sub> N/92% CO <sub>2</sub> , 68.3%(0.3 MPa)~71.6% (0.1 MPa) that of SF <sub>6</sub> <sup>[97]</sup> .
		◆ Plate-plate electrode, 0.5–1.0 MPa, 3.7% C <sub>4</sub> F <sub>7</sub> N/96.3% CO <sub>2</sub> at 0.88 MPa, 1.04 MPa reach that of SF <sub>6</sub> at 0.55 MPa, 0.65 MPa <sup>[126]</sup> .
	DC	◆ Needle-plate electrode, 0.1–0.4 MPa, 10% C <sub>4</sub> F <sub>7</sub> N/90% CO <sub>2</sub> , 80% that of SF <sub>6</sub> (positive LI), 75% that of SF <sub>6</sub> (negative LI) <sup>[102]</sup> .
		◆ Sphere-plate electrode, 0.3–0.7 MPa, 4% C <sub>4</sub> F <sub>7</sub> N/96% CO <sub>2</sub> (0.7 MPa), 8% C <sub>4</sub> F <sub>7</sub> N/92% CO <sub>2</sub> (0.7 MPa) reaches 75.05%, 81.21% that of SF <sub>6</sub> (0.5 MPa) (negative DC) <sup>[26]</sup> .
Surface flashover	◆ Needle-plate electrode, 0.3–0.7 MPa, positive DC breakdown voltage of 8% C <sub>4</sub> F <sub>7</sub> N/92% CO <sub>2</sub> is lower than 4% C <sub>4</sub> F <sub>7</sub> N/96% CO <sub>2</sub> (polarity effect) <sup>[27]</sup> .	
C <sub>5</sub> F <sub>10</sub> O	AC	◆ AC, 0.1–0.5 MPa, C <sub>4</sub> F <sub>7</sub> N/CO <sub>2</sub> with 5%, 9%, 13%, 17% C <sub>4</sub> F <sub>7</sub> N reaches 70%, 75%, 80%, and 85% of SF <sub>6</sub> <sup>[103]</sup> .
		◆ Rod-plate electrode, 0.2–0.55 MPa, C <sub>5</sub> F <sub>10</sub> O/CO <sub>2</sub> with 20 kPa, 40 kPa C <sub>5</sub> F <sub>10</sub> O reaches 61.89%, 81.99% of SF <sub>6</sub> at 0.2 MPa <sup>[127]</sup> .
	LI	◆ Sphere-sphere electrode, 0.1–0.5 MPa, C <sub>5</sub> F <sub>10</sub> O/N <sub>2</sub> and C <sub>5</sub> F <sub>10</sub> O/air with 45 kPa C <sub>5</sub> F <sub>10</sub> O reaches 84%, 91% that of SF <sub>6</sub> <sup>[128]</sup> .
		◆ Rod-plate electrode, 0.2–0.55 MPa, C <sub>5</sub> F <sub>10</sub> O/CO <sub>2</sub> with 20 kPa, 40 kPa C <sub>5</sub> F <sub>10</sub> O at 0.5 MPa reaches 88.9%, 89.9% of SF <sub>6</sub> at 0.3 MPa <sup>[128]</sup> .
	SI	◆ Plate-plate electrode, 0.1–0.2 MPa, C <sub>5</sub> F <sub>10</sub> O/air or C <sub>5</sub> F <sub>10</sub> O/CO <sub>2</sub> with 28% C <sub>5</sub> F <sub>10</sub> O is higher than that of SF <sub>6</sub> .
		◆ C <sub>5</sub> F <sub>10</sub> O/air (0.166 MPa) or C <sub>5</sub> F <sub>10</sub> O/CO <sub>2</sub> (0.114 MPa) with 10% C <sub>5</sub> F <sub>10</sub> O reaches that of SF <sub>6</sub> at 0.1 MPa <sup>[129]</sup> .
DC	◆ Needle-plate electrode, 0.1–0.5 MPa, negative SI of C <sub>5</sub> F <sub>10</sub> O/CO <sub>2</sub> with 2%–8% C <sub>5</sub> F <sub>10</sub> O is higher than that of positive, reaching 60%–70% that of SF <sub>6</sub> <sup>[130]</sup> .	
C <sub>6</sub> F <sub>12</sub> O	AC	◆ Rod-plate electrode, 0.1–0.3 MPa, C <sub>5</sub> F <sub>10</sub> O/CO <sub>2</sub> with 20 kPa C <sub>5</sub> F <sub>10</sub> O, 75%(0.3 MPa)~90%(0.1 MPa) that of SF <sub>6</sub> <sup>[131]</sup> .
		◆ Sphere-sphere electrode, 0.1–0.3 MPa, C <sub>6</sub> F <sub>12</sub> O/N <sub>2</sub> with 6% C <sub>6</sub> F <sub>12</sub> O, 1.03–1.11 times of 10% SF <sub>6</sub> /90% N <sub>2</sub> .
HFO-1234ze(E)	AC	◆ C <sub>6</sub> F <sub>12</sub> O/N <sub>2</sub> with 6% C <sub>6</sub> F <sub>12</sub> O, 1.10–1.35 times of 10% SF <sub>6</sub> /90% CO <sub>2</sub> <sup>[95]</sup> .
		◆ Sphere-plate electrode, 20% HFO1234ze(E)/80% N <sub>2</sub> , 57%–74% that of 20% SF <sub>6</sub> /80% N <sub>2</sub> ; Needle-plate electrode, 20% HFO1234ze(E)/80% N <sub>2</sub> , 92%–98% that of 20% SF <sub>6</sub> /80% N <sub>2</sub> <sup>[132]</sup> .
	LI	◆ Hemisphere-plate electrode, most of breakdowns on positive half cycle <sup>[133]</sup> .
DC	◆ Needle-plate electrode, 0.1–0.3 MPa, pure HFO1234ze(E), 74% that of SF <sub>6</sub> at 0.1 MPa, 94% of SF <sub>6</sub> at 0.3 MPa <sup>[134]</sup> .	
HFO-1336mzz(E)	AC	◆ Plate-plate electrode, pure HFO1234ze(E), 83% that of SF <sub>6</sub> <sup>[134]</sup> .
		◆ Sphere-sphere electrode, 0.1–0.3 MPa, 25% HFO-1336mzz(E)/75% CO <sub>2</sub> reaches 64%(0.1 MPa)–76%(0.2 MPa) that of SF <sub>6</sub> ; 30% HFO-1336mzz(E)/70% CO <sub>2</sub> reaches 71%(0.1 MPa)–85%(0.2 MPa) that of SF <sub>6</sub> <sup>[62]</sup> .
	DC	◆ Needle-plate electrode, 0.1–0.3 MPa, HFO-1336mzz(E)/CO <sub>2</sub> with 25%, 30% HFO-1336mzz(E) reaches 102%–105% that of SF <sub>6</sub> ; “hump” phenomenon exits with the peak at 0.2 MPa <sup>[107]</sup> .
		◆ Plate-plate electrode, 0.1–0.5 MPa, HFO-1336mzz(E)/CO <sub>2</sub> with 10%, 20% HFO-1336mzz(E) reaches 58%, 67% that of SF <sub>6</sub> ; HFO-1336mzz(E)/N <sub>2</sub> with 10%, 20% HFO-1336mzz(E) reaches 51%, 63% that of SF <sub>6</sub> <sup>[107]</sup> .

For the role of buffer gases, it is observed that the NECs of C<sub>4</sub>F<sub>7</sub>N-N<sub>2</sub> and C<sub>4</sub>F<sub>7</sub>N-CO<sub>2</sub> arcs are very close to that of C<sub>4</sub>F<sub>7</sub>N arc until the buffer gas proportion reaches 50% (Figure 6(d))<sup>[40]</sup>. In general, the works on calculating radiation coefficients of eco-friendly

gaseous arcs are not as much as those on the thermodynamic and transport coefficients. One of the likely explanations is the lack of atomic and molecular spectrum data and the cross sections of collision processes.



**Fig. 6 Basic data of eco-friendly gaseous arcs.** (a) Particle compositions. Reprinted with permission from Ref. [141], © 2021 Wiley-VCH GmbH. (b) Thermodynamic properties. Reprinted with permission from Ref. [142], © 2019 Springer Science Business Media, LLC, part of Springer Nature. (c) Transport coefficients. Reprinted with permission from Ref. [140], © 2018 AIP Publishing. (d) Radiation coefficients. Reprinted with permission from Ref. [142], © 2019 Springer Science Business Media, LLC, part of Springer Nature. (e) Dielectric breakdown properties. Reprinted with permission from Ref. [160], © 2015, EDP Sciences, SIF, Springer-Verlag Berlin Heidelberg. (f) Arc decaying characteristics. Reprinted with permission from Ref. [142], © 2019 Springer Science Business Media, LLC, part of Springer Nature.

### Post-arc dielectric breakdown properties

During the arc interruption process, a high electric field strength caused by a transient recovery voltage (TRV) is applied to the hot gaseous medium between electrodes. If the hot gas has a lower dielectric strength than the applied electric field strength, the gas breakdown will occur, leading to the re-ignition of the arc<sup>[155]</sup>. In this case, the insulating gases remain a much higher temperature than room temperature during the arc extinction<sup>[156]</sup>. It is therefore necessary to investigate the dielectric breakdown properties of hot gas mixtures rather than cold gases at room temperature. This is usually achieved by the analysis of Boltzmann equation which describes the electron transport in hot gases. Bolsig+ is the widely used toolkit for solving the Boltzmann equation of weakly ionized gases<sup>[157]</sup>. In addition, the particle compositions at the temperatures considered and the electron-impact cross sections including elastic, ionization, electron attachment and excitation cross sections are needed in the calculation<sup>[155, 156, 158]</sup>. Most previous works on this topic are devoted to SF<sub>6</sub> and its various mixtures<sup>[156, 159, 160]</sup>. Very few works focus on eco-friendly gases, except for C<sub>3</sub>F<sub>8</sub><sup>[158]</sup>. It is found that hot C<sub>3</sub>F<sub>8</sub> gas has much poorer dielectric breakdown performance than SF<sub>6</sub> (Figure 6(e))<sup>[158]</sup>.

### Evaluation of arc-quenching performance

The arc-quenching performance is usually evaluated by means of numerical modeling or experiments. The former approach is generally cheaper than the latter one. Based on the thermodynamic properties, transport coefficients, and radiation coefficients, a computational fluid model, namely magnetohydrodynamics (MHD) model can be established to describe arc characteristics and thus evaluate arc interruption ability. CO<sub>2</sub> has been investigated by the MHD modeling that it has a lower arc core temperature and larger arc radius than SF<sub>6</sub><sup>[161]</sup>. Using the same method, the arc-quenching performance of C<sub>4</sub>F<sub>7</sub>N-CO<sub>2</sub> and C<sub>5</sub>F<sub>10</sub>O-CO<sub>2</sub> mixtures has been compared with pure SF<sub>6</sub> and CO<sub>2</sub> (Figure 6(f))<sup>[162]</sup>. Considering that 2-D and 3-D MHD modeling are always time-con-

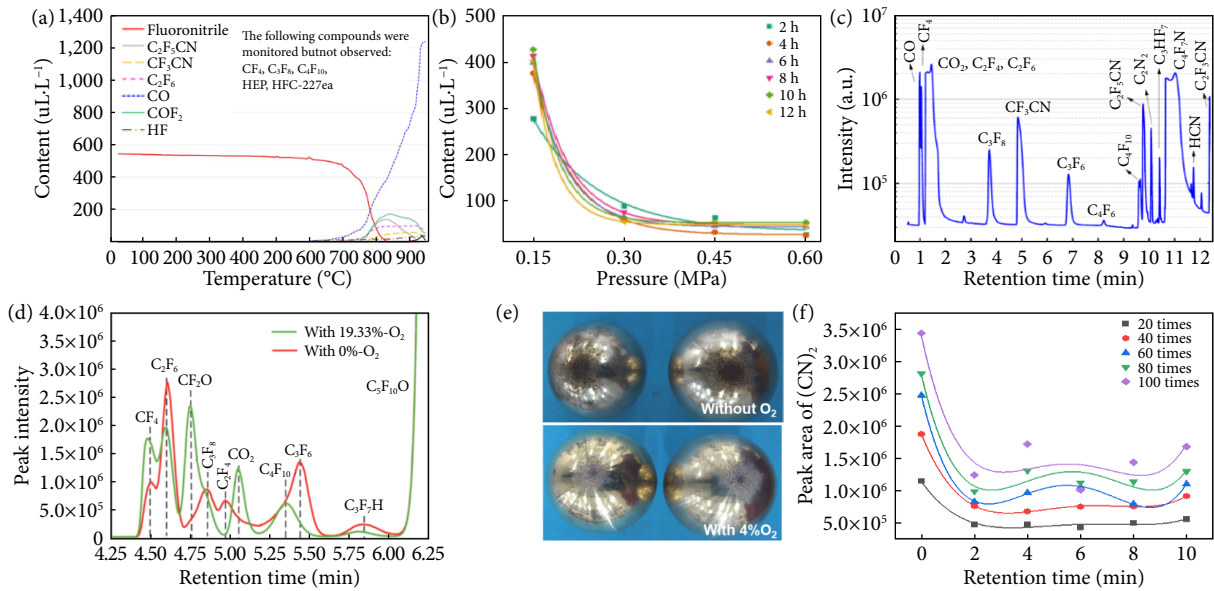
suming, 1-D modeling is used in investigating the decaying characteristics of CF<sub>3</sub>I and C<sub>4</sub>F<sub>7</sub>N arcs<sup>[140]</sup>. For instance, the result of the 1-D arc model indicates that CF<sub>3</sub>I shows a higher performance of thermal interruption than CO<sub>2</sub>, N<sub>2</sub> and air<sup>[163]</sup>. To further reveal the arc interruption ability in a more sophisticated way, a fast evaluation method is proposed by combining 1-D hydrokinetic modeling and Boltzmann equation analysis<sup>[135, 164]</sup>. However, due to the lack of electron-impact cross sections, this method is only applied to evaluate the arc-quenching performance of SF<sub>6</sub>, CF<sub>4</sub>, CO<sub>2</sub>, and air. In addition to numerical works, some researchers performed experimental investigations on the arc-quenching of eco-friendly gases by measuring arc voltage, current, and emission spectrum. They found that C<sub>5</sub>F<sub>10</sub>O-CO<sub>2</sub> arc is more volatile than SF<sub>6</sub> gas, and adding C<sub>5</sub>F<sub>10</sub>O into CO<sub>2</sub> can improve the stability of the arc, and significantly reduces the arc temperature<sup>[165]</sup>. C<sub>4</sub>F<sub>7</sub>N and its mixtures with CO<sub>2</sub> are also tested experimentally in a disconnector, and the results show that the arcing time is stable over 100 operations<sup>[96]</sup>.

### 3.3 Stability and decomposition

Eco-friendly insulating gas will decompose to low carbon or fluorine fractions under discharges or over-thermal faults. These products have inferior dielectric strength and will not fully recombine to the original molecules, which may severely decrease the insulation and interruption capacity of gas. Therefore, the decomposition characteristics of eco-friendly gas is important in evaluating the operating feasibility, which has been explored in thermal, discharge aspects as well as theoretical level.

#### Thermal decomposition

The current heat effect or poor contact induced partial over-thermal fault (POF) are the main reason for thermal decomposition. Recent studies have clarified the thermal decomposition conditions, by-product type and content of eco-friendly insulating gas. For example, the initial decomposition temperature of pure gaseous C<sub>4</sub>F<sub>7</sub>N (test conducted based on Tubular furnace) reaches 500–650°C, and CF<sub>3</sub>CN, C<sub>3</sub>F<sub>6</sub>, C<sub>2</sub>F<sub>5</sub>CN are detected by Fourier



**Fig. 7** The thermal and discharge decomposition properties of eco-friendly insulating gas. (a) Decomposition products from thermal degradation of C<sub>4</sub>F<sub>7</sub>N. Reprinted with permission from Ref. [57], © 2016 IEEE. (b) The C<sub>3</sub>F<sub>6</sub> generated by C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> thermal decomposition at 450°C under different pressures. Reprinted with permission from Ref. [169], © 2020 Author(s). (c) The decomposition products of C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> after multiple breakdowns. Reprinted with permission from Ref. [172], © 2019 AIP Publishing. (d) The decomposition products of C<sub>5</sub>F<sub>10</sub>O/N<sub>2</sub>/O<sub>2</sub> after multiple breakdowns. Reprinted with permission from Ref. [181], © 2019 Author(s). (e) The electrode morphology of C<sub>4</sub>F<sub>7</sub>N/N<sub>2</sub> gas mixture after multiple breakdowns. Reprinted with permission from Ref. [182], © 2019 IEEE. (f) The influence of O<sub>2</sub> content on the yield of C<sub>2</sub>N<sub>2</sub>. Reprinted with permission from Ref. [182], © 2019 IEEE.

infrared spectrometer (FTIR) or gas chromatography-mass spectrometer (GC-MS)<sup>[56,166,167]</sup>. Other by-products such as CO, C<sub>2</sub>F<sub>4</sub>, C<sub>2</sub>F<sub>6</sub>, C<sub>3</sub>F<sub>8</sub> and C<sub>2</sub>N<sub>2</sub> are generated at higher temperature (700°C) (Figure 7(a))<sup>[56]</sup>. The utilization of metal thermocouples for POF simulation obtained lower initial decomposition temperature (~350°C), which is ascribed to the participation of the metal interface<sup>[167]</sup>. Specifically, C<sub>3</sub>F<sub>6</sub> is first detected, followed by C<sub>2</sub>N<sub>2</sub>, CF<sub>4</sub>, C<sub>3</sub>F<sub>8</sub>, etc. The fluorinated ketones demonstrate similar thermal decomposition properties, and the by-products including CF<sub>4</sub>, C<sub>2</sub>F<sub>6</sub>, C<sub>3</sub>F<sub>8</sub>, C<sub>3</sub>F<sub>6</sub>, C<sub>3</sub>F<sub>7</sub>H, C<sub>5</sub>F<sub>12</sub> are generated<sup>[168]</sup>.

Besides, the gas pressure plays an important role in thermal decomposition. The amount of various decomposition products decreases with the increase of gas pressure and tends to be stable at ~0.3 MPa (Figure 7(b))<sup>[167]</sup>. This is because the increase of gas dissociation reaction rate constant is less than linear with the pressure<sup>[166]</sup>. Thus, the thermal stability of eco-friendly insulating gas at higher pressure is superior, which favors the HV-GIE application. Besides, the addition of >6% O<sub>2</sub> demonstrates a negative impact on thermal stability<sup>[169]</sup>. The formation of C<sub>3</sub>F<sub>8</sub>, C<sub>3</sub>F<sub>6</sub>, CF<sub>3</sub>CN, C<sub>2</sub>F<sub>5</sub>CN and C<sub>2</sub>N<sub>2</sub> is inhibited while the content of CF<sub>4</sub>, COF<sub>2</sub> increases. Further investigation on the influence of trace impurities (such as H<sub>2</sub>O) on the thermal stability and the gas aging mechanism under electrothermal conditions is necessary.

### Discharge decomposition

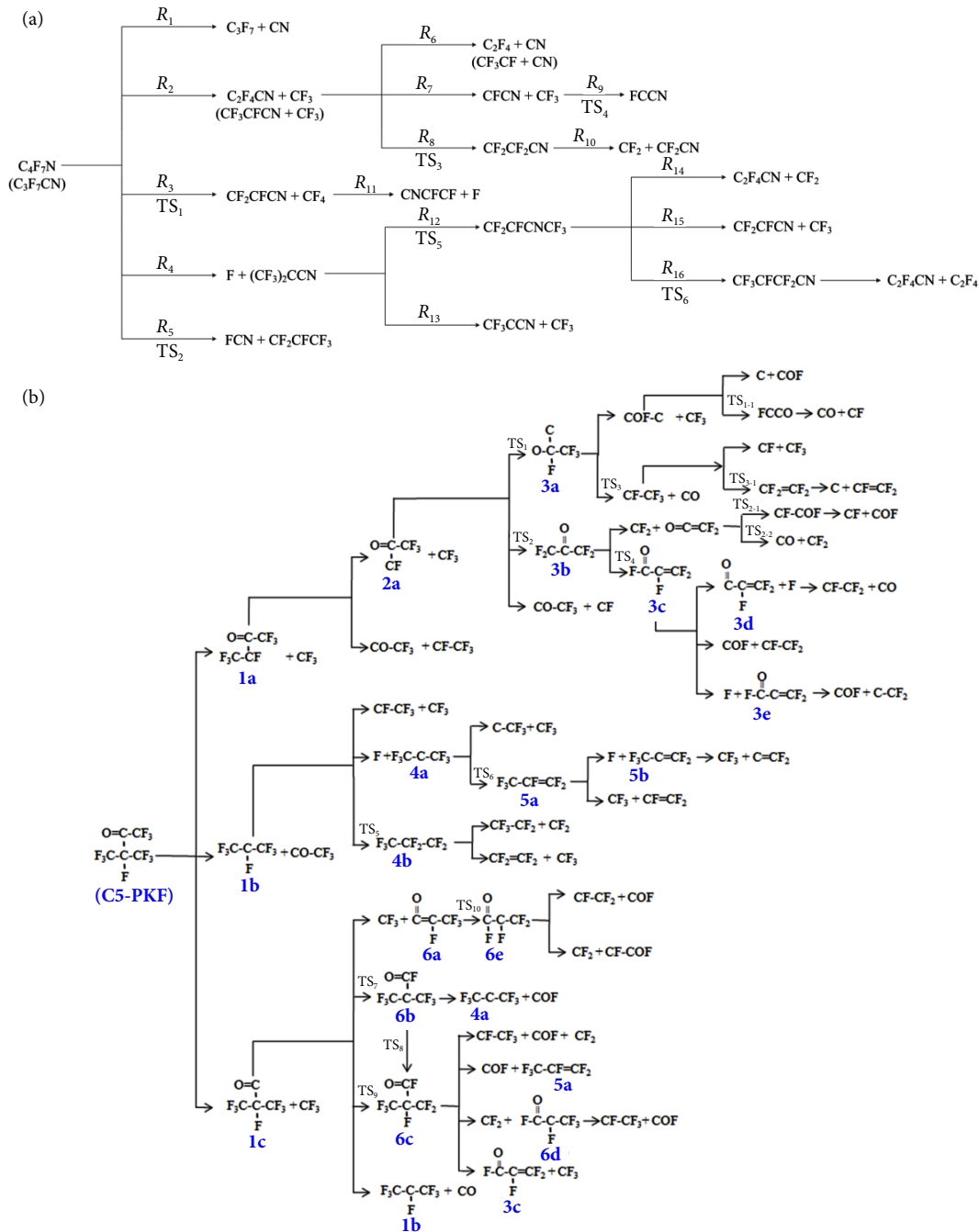
The discharge decomposition of eco-friendly insulating gas is mainly caused by collision ionization, photoionization and thermal ionization, and various fractions (positive, negative ions, neutral particles) will be generated. Meanwhile, the fractions will react with each other to form decomposition by-products. The PD, dielectric breakdown, arc-quenching, etc. cause decomposition of eco-friendly gases. For C<sub>4</sub>F<sub>7</sub>N gas mixture, common by-products including CO, CF<sub>4</sub>, C<sub>2</sub>F<sub>6</sub>, C<sub>2</sub>F<sub>4</sub>, C<sub>3</sub>F<sub>8</sub>, C<sub>3</sub>F<sub>6</sub>, CF<sub>3</sub>CN, C<sub>2</sub>N<sub>2</sub>, C<sub>2</sub>F<sub>5</sub>CN, C<sub>4</sub>F<sub>8</sub> are detected (Figure 7(c))<sup>[170,171]</sup>. For fluorinated ketones, CO<sub>2</sub>, CF<sub>4</sub>, C<sub>2</sub>F<sub>6</sub>, C<sub>2</sub>F<sub>4</sub>, C<sub>3</sub>F<sub>8</sub>, C<sub>3</sub>F<sub>6</sub>, C<sub>3</sub>F<sub>7</sub>H, C<sub>4</sub>F<sub>10</sub>, C<sub>5</sub>F<sub>12</sub>, C<sub>6</sub>F<sub>14</sub>, C<sub>4</sub>F<sub>8</sub> are

generated (Figure 7(d))<sup>[172–179]</sup>. The dielectric strength of most decomposition by-products is inferior to that of electron affinity gas, and the C<sub>4</sub>F<sub>8</sub>, CF<sub>3</sub>CN, C<sub>2</sub>F<sub>5</sub>CN, C<sub>2</sub>N<sub>2</sub>, C<sub>3</sub>F<sub>6</sub>, COF<sub>2</sub>, HF, HCN belongs to corrosive or toxic substances, which may pose a potential threat to insulation stability and maintenance personal safety.

Importantly, current experiments confirm that the generated fractions cannot fully recombine to electron affinity gas itself after discharge, which is ascribed to the complex molecular structure with low symmetry. There also exists a “cumulative effect” of the generated by-products. That is, the yield of various by-products shows a linear growth trend with the discharge intensity, period, and the electron affinity gas in the mixture is constantly consumed. The mixing ratio should be monitored to avoid the impact of insulation degradation caused by continuous consumption of electron affinity gas on GIE operation reliability. Besides, the formation or participation of solid by-product exists for fluorocarbon eco-friendly insulating gas, especially when N<sub>2</sub> is employed as the buffer gas (Figure 7(e))<sup>[180]</sup>. The addition of O<sub>2</sub> is capable of inhibiting the most gaseous and solid decomposition by-products (Figure 7(f))<sup>[180–182]</sup>. On one hand, O<sub>2</sub> has a preferable arc-quenching capability than CO<sub>2</sub> or N<sub>2</sub>, which causes faster decaying rate of arc conductivity<sup>[181,182]</sup>. On the other, the carbon particles generated by arc discharge can be oxidized to CO or CO<sub>2</sub> by O<sub>2</sub> during discharge<sup>[182]</sup>. It should be noticed that higher content of O<sub>2</sub> brings negative effect on gas stability. It is necessary to establish the correlation between discharge and decomposition properties, as well as to explore the suppression scheme of by-products to further improve the stability of eco-friendly insulating gas.

### Decomposition mechanism

Understanding the decomposition mechanism is significant for gas performance optimization. The development of quantum chemistry and multi-physics simulation promotes relevant research. The generation/loss rate of by-product is determined by chemical reactions and the initial concentration according to mass action law, which should be investigated in chemical kinetics

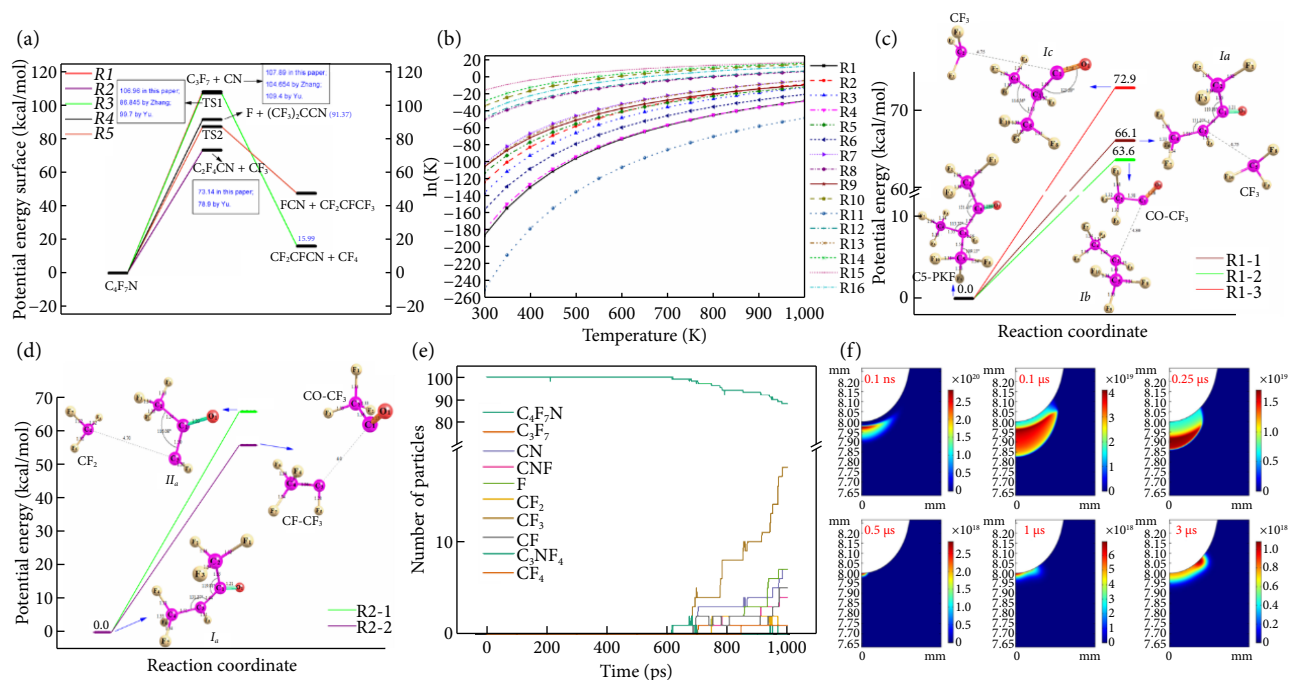


**Fig. 8** The decomposition scheme and reaction pathways of eco-friendly insulating gas. (a)  $C_4F_7N$ . Reprinted with permission from [185], © 2019 IOP Publishing. (b)  $C_5F_{10}O$ . Reprinted with permission from [187], © 2016 Author(s).

model with a complete set of reactions. The relatively-complete decomposition scheme and reaction pathways of eco-friendly gases (such as  $C_4F_7N$  and  $C_5F_{10}O$ ) are shown in Figure 8<sup>[183–185]</sup>. The potential energy surface of various reactions can be obtained by DFT and transition state (TS) calculations (Figures 9(a)–9(d)). For  $C_4F_7N$ , a slight barrier height in reaction  $R_1$  ( $C_4F_7N \rightarrow C_3F_7 + CN$ ) exists, which is less likely to occur than reaction  $R_2$  ( $C_4F_7N \rightarrow C_2F_4CN + CF_3$ ) and  $R_4$  ( $C_4F_7N \rightarrow F + (CF_3)_2CCN$ ). The reaction  $R_5$  ( $C_4F_7N \rightarrow TS_2 \rightarrow FCN + CF_2CFCF_3$ ) is more likely to occur than reaction  $R_3$  in  $C_4F_7N$  dissociation (Figure 9(a)). Besides,  $R_5$  is the most important reaction leading to the dissociation of  $C_4F_7N$  below 600 K with larger reaction constants, while reaction  $R_2$  takes the place of reaction  $R_5$  over 600 K to 3,300 K and reaction  $R_3$  become dominant above 700 K (Figure 9(b))<sup>[183]</sup>. For  $C_5F_{10}O$ , the

favorable decomposition reactions include C–C bond ruptures producing **1a** and **1b**, followed by subsequent C–C bond ruptures and internal F atom transfers in the decomposition of **1a** and **1b**, respectively (Figures 9(c) and 9(d))<sup>[185]</sup>.

Moreover, chemical kinetic models including local thermodynamic equilibrium (LTE) and non-local chemical equilibrium (non-LCE) can be used to explore the gas composition, as summarized in Section 3.2. The ReaxFF molecular dynamics (ReaxFF-MD) is also proposed to explore the decomposition process of eco-friendly gas<sup>[186–190]</sup>. The bond cleavage and formation during chemical reactions are described by the ReaxFF force field, which can be optimized by DFT<sup>[187]</sup>. The main particle including  $CF_3$ ,  $CF_2$ ,  $CF$ ,  $CN$ ,  $F$ ,  $CNF$ ,  $C_3F_7$ ,  $C_3NF_4$ ,  $C_4NF_6$ ,  $CF_4$  etc. are generated in  $C_4F_7N$ ,  $C_5F_{10}O$ ,  $C_6F_{12}O$  gas mixture (Figure 9(e))<sup>[186–190]</sup>. Besides, the



**Fig. 9** The decomposition mechanism of eco-friendly insulating gas. (a) Decomposition potential energy surface of  $C_4F_7N$  including reactions  $R_1$ – $R_5$ . Reprinted with permission from Ref. [185], © 2019 IOP Publishing. (b) Rate constants of  $C_4F_7N$  decomposition reactions during 300–1,000 K. Reprinted with permission from Ref. [185], © 2019 IOP Publishing. (c) Decomposition potential energy surface of  $C_5F_{10}O$  (generating Ia and Ib). Reprinted with permission from Ref. [187], © 2016 Author(s). (d) Decomposition potential energy surface of Ia. Reprinted with permission from Ref. [187], © 2016 Author(s). (e) Time evolution of the  $C_4F_7N$  decomposed major species at 1,900 K. Reprinted with permission from Ref. [188], © 2017 Author(s). (f) Distribution of positive ion density near the discharge electrode in  $C_4F_7N/CO_2$ . Reprinted with permission from Ref. [193], © 2022 Author(s).

numerical simulation model of chemical kinetics–fluid is proposed to explore the PD decomposition mechanism insulating gas. For  $C_4F_7N$ , the 2D axisymmetric model combines the drift-diffusion and Poisson's equations were used and the distribution of positive, negative ions and electrons were obtained (Figure 9(f))<sup>[191]</sup>. Further research on the regulation of decomposition by-products generation and inhibition of solid precipitation mechanism needs to be clarified based on the above-mentioned schemes.

### 3.4 Materials compatibility

Assessment on the materials compatibility of eco-friendly gas has been focused on metals, epoxy resin, elastomers, adsorbent (or desiccant) currently. The aging tests were conducted at specific period or temperature, followed by the gas composition and solid materials properties analysis<sup>[192,193]</sup>. Besides, DFT or molecular dynamics (MD) studies were conducted to further reveal the gas–solid interface interaction mechanism<sup>[194]</sup>.

#### Metal

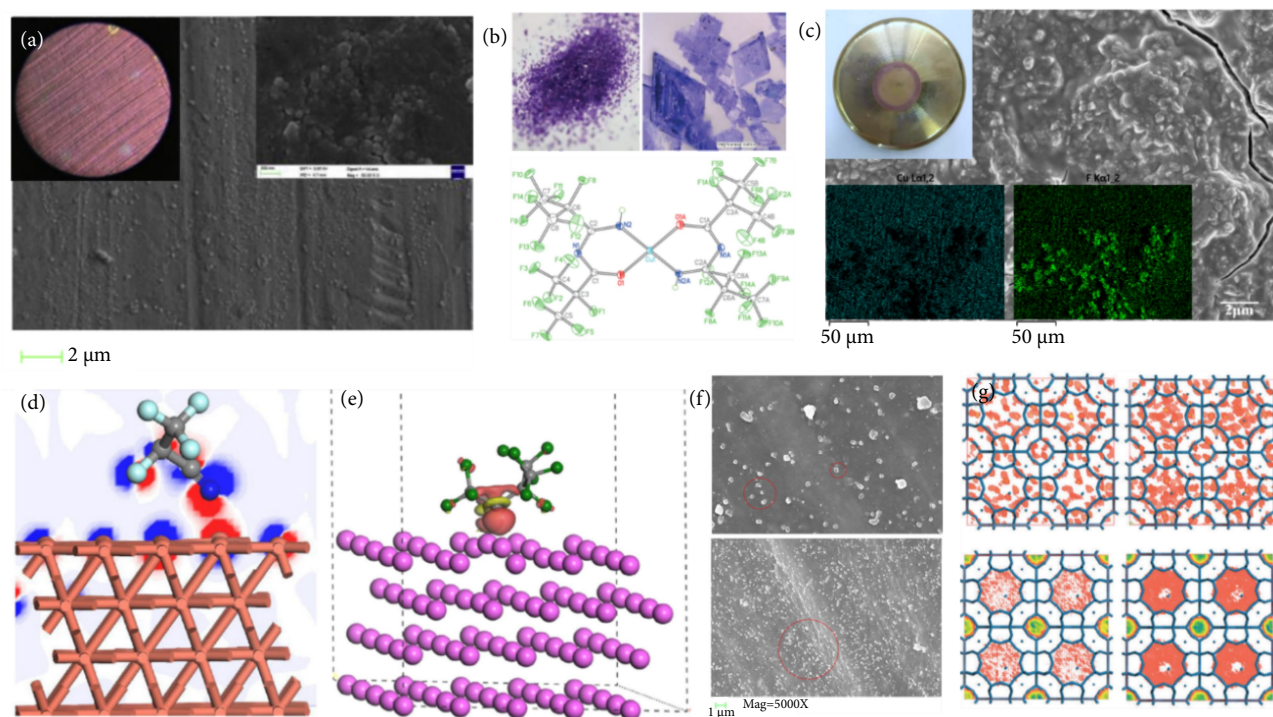
The compatibility between fluorinated-nitrile, ketones, and HFO-1234ze(E) with copper, aluminum, silver has been evaluated. Copper demonstrated poor stability when contacted with  $C_4F_7N$ ,  $C_5F_{10}O$ , and  $C_6F_{12}O$  at 120–220°C<sup>[195–200]</sup>. The copper surface was corroded with fluoride accumulation, and the generation of  $C_3F_6$  and  $C_3F_7H$  was found (Figure 10(a))<sup>[201–203]</sup>. Further analysis confirmed that the square planar copper(II) complex with two per-fluorinated N-acylamidine ligands was formed during the multi-step interface reaction(Figure 10(b))<sup>[204]</sup>. The interaction between fluorinated-nitrile, ketones and aluminum, silver had little influence while the formation of  $C_3F_6$  existed at high interface temperature. Besides, the PD induced decomposition also results in copper corrosion or fluorocarbon solid by-product precipitation, as shown in

Figure 10(c)<sup>[204]</sup>.

The interaction mechanism of fluorinated nitrile, ketone with metal is also investigated. The interaction energy and electronic parameters such as charge transfer, density of states were revealed based on DFT. Specifically, relevant evaluations on the Cu, Al, Ag, Zn, and ZnO surfaces confirmed the strong interaction activity of  $-CN$ , and  $-C=O$  group of  $C_4F_7N$ ,  $C_5F_{10}O$  and  $C_6F_{12}O$ . The obvious electron charge transfer and bonding were found (Figures 10(d) and 10(e))<sup>[205,206]</sup>. The adsorption-dissociation process was also explored, indicating the metal acts as the catalytic interface for gas decomposition. The formation of  $C_3F_6$  might be ascribed to this. Overall, anti-corrosion treatment of copper is needed for fluorinated-nitrile, ketone based GIE. Future exploration on the metal alloy, anti-corrosion coating as well as the impact of gas-metal interaction process on the electrical, thermal conductivity should be clarified.

#### Epoxy resin

Epoxy resin is normally used as solid insulating material in GIE. The compatibility assessment on  $C_4F_7N/CO_2$  found that the DC conductivity of epoxy resin decreased with the aging temperature, while AC conductivity demonstrates an increasing trend<sup>[207]</sup>. The surface flashover voltage was kept stable after interaction. Higher aging temperature (>160°C) resulted in the formation of  $C_3F_6$  and  $C_{12}F_{21}N_3$ . The  $C_4F_7N$ -epoxy resin interface discharge decomposition properties were also investigated<sup>[208,209]</sup>. Various by-products including  $CF_4$ ,  $C_3F_8$ ,  $C_6F_{14}$ ,  $C_3F_6$ ,  $C_4F_6$ ,  $C_4F_8$ ,  $CF_3CN$ , and  $C_{12}F_7H_{17}O_2$  were detected, which content increased with the PD period. Moreover,  $-CF$  and  $-CN$  were found on the epoxy resin surface. Further explorations on the interaction properties between eco-friendly gas and the main decomposition by-products should be conducted.



**Fig. 10** The materials compatibility of eco-friendly insulating gas. (a) The morphology of copper after interacted with C<sub>4</sub>F<sub>7</sub>N at 220°C for 40 h. Reprinted with permission from Ref. [202], © 2019 Elsevier Ltd. (b) Microscope photograph of violet crystals and molecular structure of square planar copper(II) complex. Reprinted with permission from Ref. [206], © 2020 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (c) The morphology of copper electrode after 96 h PD (15% C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> gas mixture, 0.15 MPa) Reprinted with permission from Ref. [206], © 2020 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (d) The electron density difference of C<sub>4</sub>F<sub>7</sub>N-Cu(1 1 1) interface. Reprinted with permission from Ref. [207], © 2018 Elsevier Ltd. (e) The electron density difference of C<sub>5</sub>F<sub>10</sub>O-Cu(1 1 1) interface. Reprinted with permission from Ref. [208], © 2019 Elsevier Ltd. (f) The morphology of EPDM after interacted with C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub>. Reprinted with permission from Ref. [213], © 2021 Author(s). (g) The density fields of HF(up) and CO(down) in the Na-4A zeolite under the pressures of 0.01, 1 kPa. Reprinted with permission from Ref. [218], © 2020 Author(s).

### Elastomer

Elastomer or rubber ring plays an important role in GIE considering the relatively high working pressure. At present, assessment on the compatibility between fluorinated-nitrile, ketone with ethylene propylene diene monomer (EPDM), nitrile butadiene rubber (NBR), fluoro rubber has confirmed the incompatibility of EPDM and NBR<sup>[210–212]</sup>. Specifically, the C<sub>4</sub>F<sub>7</sub>N-EPDM interaction induced the generation of CO, C<sub>3</sub>F<sub>6</sub>, C<sub>2</sub>H<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, the crystal precipitation and mechanical degradation of rubber (Figure 10(f))<sup>[210]</sup>. Moreover, the ethylidene norbornene (ENB)-EPDM demonstrated superior gas-solid stability compared to the dicyclopentadiene (DCPD-EPDM), and the silicone grease coating could further prevent the interaction<sup>[211]</sup>. Similar studies on the NBR and C<sub>4</sub>F<sub>7</sub>N confirmed the mechanical properties deterioration with 10.15% decrease of elongation at break and 37% loss of tensile strength<sup>[212]</sup>.

In addition, the sealing performance of buffer gas with a smaller molecular structure should be considered. For example, butyl rubber is tight to CO<sub>2</sub>, air, or N<sub>2</sub>, while the unsaturated double bonds in the isoprene chain might be oxidized by the discharge induced O<sub>3</sub>. The utilization of halogenated butyl rubber (CIIR-BIIR) with a similar molecular structure is recommended. Relevant gas permeability tests confirmed CIIR-BIIR demonstrated a lower permeation rate coefficient than that of EPDM<sup>[56]</sup>. Further explanations on the design of special sealing rubber for eco-friendly insulating gas are necessary.

### Adsorbent

Adsorbent or desiccant is mainly utilized for impurities adsorption

especially trace moisture in SF<sub>6</sub>-based GIE. For eco-friendly insulating gas, the additional capabilities such as the elimination of toxic and corrosive products should be considered as the “cumulative effect” of decomposition by-products. Evaluation of the compatibility of γ-Al<sub>2</sub>O<sub>3</sub>, molecular sieve (3A–5A) found that there existed strong interaction between γ-Al<sub>2</sub>O<sub>3</sub> and C<sub>4</sub>F<sub>7</sub>N, causing consumption of the main insulating gas<sup>[213–215]</sup>.

As for the by-products, it was found that molecular sieves demonstrated a weak adsorption effect on PFCs (CF<sub>4</sub>, C<sub>2</sub>F<sub>6</sub>, C<sub>3</sub>F<sub>8</sub>) and CO. The 3A–5A molecular sieve is capable of removing C<sub>2</sub>N<sub>2</sub>, CF<sub>3</sub>CN<sup>[215]</sup>. The microscopic parameters including adsorption isotherm, saturated adsorption capacity, free energy, diffusion coefficient, etc. of gas-adsorbent system were also calculated based on MD, and the superior adsorption performance of HF, COF<sub>2</sub>, C<sub>2</sub>N<sub>2</sub>, CF<sub>3</sub>CN, C<sub>2</sub>F<sub>5</sub>CN was confirmed (Figure 10(g))<sup>[216]</sup>. The design and synthesis of new adsorbent is highly recommended for the eco-friendly GIE application<sup>[217]</sup>. The metal organic frameworks (MOFs) with periodic network structures formed by molecular self-assembly of metal ions or ion clusters and organic ligands is a promising selection.

### 3.5 Biosafety

Biosafety is another significant parameter for eco-friendly insulating gas considering the inevitable exposure of maintenance personnel. Some toxicity properties such as acute toxicity, target organ toxicity have been investigated currently.

### LC50

The Material Safety Data Sheet (MSDS) of C<sub>4</sub>F<sub>7</sub>N and C<sub>5</sub>F<sub>10</sub>O

given by 3M indicated that the median lethal concentration (LC<sub>50</sub>, 4 h, rat) of C<sub>4</sub>F<sub>7</sub>N, C<sub>4</sub>F<sub>7</sub>N was 10,000–15,000 μL·L<sup>-1</sup> (ppm), 14,000–20,000 μL·L<sup>-1</sup>, respectively (Figure 11(a)), and the no observed adverse effect level (NOAEL) of 28 days subacute inhalation of C<sub>4</sub>F<sub>7</sub>N in rats was 500 μL·L<sup>-1</sup>. The LC<sub>50</sub> (4 h, mice) of C<sub>4</sub>F<sub>7</sub>N given by Wuhan University was 1175 μL·L<sup>-1</sup> (male) and 1,380 μL·L<sup>-1</sup> (female), the result given by Schneider was 1600 μL·L<sup>-1</sup>, the difference in LC<sub>50</sub> values was due to the difference in species and weight of the mice used<sup>[218,219]</sup>. The occupational exposure limits (OEL) of C<sub>4</sub>F<sub>7</sub>N and C<sub>5</sub>F<sub>10</sub>O were set to 65 μL·L<sup>-1</sup> and 225 μL·L<sup>-1</sup>, which is lower than 1000 μL·L<sup>-1</sup> of SF<sub>6</sub><sup>[218,219]</sup>. For the C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> gas mixture, the LC<sub>50</sub> (rat, 4 h) of 4% C<sub>4</sub>F<sub>7</sub>N-96% CO<sub>2</sub> and 10% C<sub>4</sub>F<sub>7</sub>N-90% CO<sub>2</sub> gas mixture reaches 16%–21.1% and 9.55%–10%<sup>[56]</sup>. Considering the content of C<sub>4</sub>F<sub>7</sub>N in the gas mixture is generally in the range of 4%–10%, the C<sub>4</sub>F<sub>7</sub>N gas mixture is classified as non-toxic substance according to EU 1272/2008 CLP and Globally Harmonized System (GHS) regulation<sup>[56]</sup>.

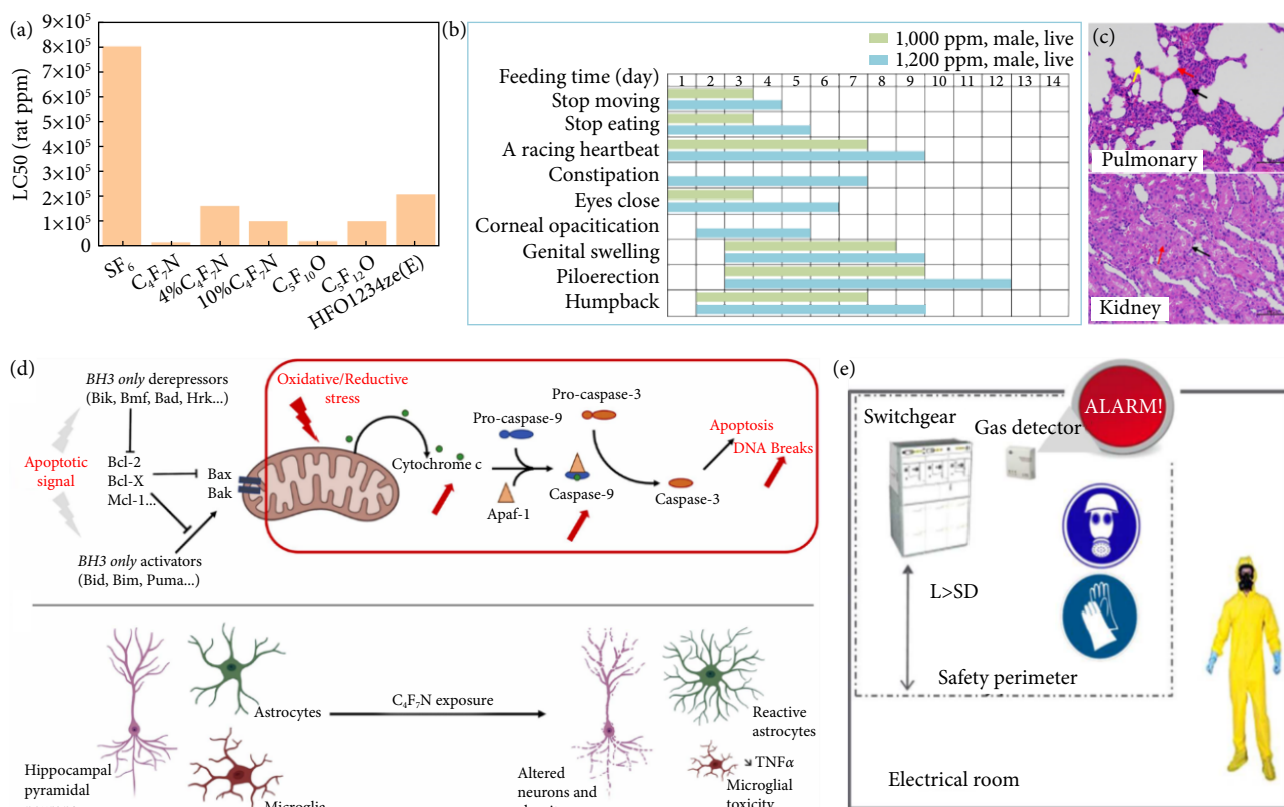
At present, the acute toxicity of C<sub>4</sub>F<sub>7</sub>N or C<sub>5</sub>F<sub>10</sub>O has been preliminarily confirmed to be higher than SF<sub>6</sub> and other eco-friendly gases such as HFO-1234ze(E). There is a lack of in-depth understanding of the inhalation toxicity of C<sub>4</sub>F<sub>7</sub>N or C<sub>5</sub>F<sub>10</sub>O, especially the hazard mechanism to individual vital signs under different exposure cycles (acute, subacute, and chronic).

#### Target organ toxicity

The preliminary study on the vital signs changing and target organ toxicity of C<sub>4</sub>F<sub>7</sub>N found that rats or mice would suffer from

gloomy spirit, action retardation and abnormal blood cell count after exposure (Figure 11(b)). The pathological tissue slice further confirmed the lung, kidney, intestine and brain injury when the exposure content reaches 1.5%, as shown in Figure 11(c)<sup>[220,221]</sup>. Besides, the behavioral experiments (spontaneous alternation in the Y-maze and object recognition) on mice that were exposed to 800 μL·L<sup>-1</sup> and 1500 μL·L<sup>-1</sup> C<sub>4</sub>F<sub>7</sub>N indicated the general impact on mobility, anxiety, spatial working memory and recognition memory. The behavioral impairments occurred on day 1, day 7 and object recognition on day 14. The histology and immunohistochemistry tests of the brain further observed histological alterations of pyramidal neuronal layer in the hippocampus, neuroinflammatory astroglial reaction, and microglial alterations<sup>[222]</sup>. As illustrated in Figure 11(d), C<sub>4</sub>F<sub>7</sub>N could result in a significant increase in γ-H2Ax/H2Ax levels ratio, and cell loss in the hippocampus, triggering proapoptotic signals including apoptosis, DNA breaks. The gas exposure also causes cellular impact, inducing neuroinflammation and microglia damage.

Considering the potential lung, kidney and brain injury caused by C<sub>4</sub>F<sub>7</sub>N, the biochemical specific indicators, safety concentration threshold of short-term and long-term exposure should be determined. The utilization of effective personal protective equipment (PPE) is necessary for risk-exposed personnel (Figure 11(e)). Further exploration of reproductive, genetic toxicity of C<sub>4</sub>F<sub>7</sub>N and other eco-friendly insulating gas should be conducted to guide safety application.



**Fig. 11** The biosafety of eco-friendly insulating gas. (a) The LC<sub>50</sub> (rat, 4 h) of SF<sub>6</sub> and various eco-friendly insulating gas. (b) The changes in vital signs of mice that survived exposure to different concentrations of the gas over a 14-day observation period. Reprinted with permission from Ref. [223], © 2020 Taylor & Francis. (c) Pathological section of the pulmonary and kidney after 2% C<sub>4</sub>F<sub>7</sub>N exposure for 4 h. Reprinted with permission from Ref. [222], © 2019 Elsevier Ltd. (d) Summary of the toxicity observed in the mice brains after exposure to C<sub>4</sub>F<sub>7</sub>N (up) and the impact on brain cells (down). Reprinted with permission from Ref. [224], © 2022 Elsevier Ltd. (e) Representation of an electrical room with a safety perimeter around the electrical switchgear when gas leakage is detected. Reprinted with permission from Ref. [232], © 2018 Author(s).



### By-products toxicity

The generation of various toxic by-products is inevitable for GIE, especially for arc-quenching or under fault conditions. The highly toxic H<sub>2</sub>S, SO<sub>2</sub>, SOF<sub>2</sub>, HF, SO<sub>2</sub>F<sub>2</sub> is found in SF<sub>6</sub> GIE. For eco-friendly insulating gas such as C<sub>4</sub>F<sub>7</sub>N, the 100 times breaking tests on the 145 kV circuit breaker with 6% C<sub>4</sub>F<sub>7</sub>N-89.1% CO<sub>2</sub>-4.9% O<sub>2</sub> under 40 kA short circuit current possessed the acute inhalation toxicity (LC50) of 3.8%, lower than 7.4% of SF<sub>6</sub><sup>[223]</sup>. The 200 times breaking tests on the load switch filled with 57% C<sub>4</sub>F<sub>7</sub>N-43% air at 630 A resulted in the LC50 of 31 μL·L<sup>-1</sup> (male mice) and 34 μL·L<sup>-1</sup> (female mice), respectively. The most toxic by-product was perfluoroisobutylene (C<sub>4</sub>F<sub>8</sub>) with the LC50 of 0.5 μL·L<sup>-1</sup>, followed by C<sub>2</sub>N<sub>2</sub> (175 μL·L<sup>-1</sup>), COF<sub>2</sub> (180 μL·L<sup>-1</sup>), CF<sub>3</sub>CN (250 μL·L<sup>-1</sup>), and CO (1,880 μL·L<sup>-1</sup>)<sup>[224]</sup>. In addition, the 42% C<sub>5</sub>F<sub>10</sub>O-58% Air gas mixture after 100 breakings at 630 A demonstrated the LC50 of 21,000 μL·L<sup>-1</sup>, which was much higher than SF<sub>6</sub> under the same conditions<sup>[225]</sup>.

Overall, the acute inhalation toxicity of the gas mixture after arc-quenching significantly changed due to the formation of various toxic by-products, which is determined by the content of main insulating gas, number of operations and breaking capacity. There are few studies on the biosafety of eco-friendly insulating gas under the long-term operation of POF, PD fault conditions. The development of a selective adsorbent or desiccant that can filter out highly toxic decomposition by-products to further improve the application safety of eco-friendly insulating gas-based GIE is urgent.

## 4 Application of eco-friendly insulating gas

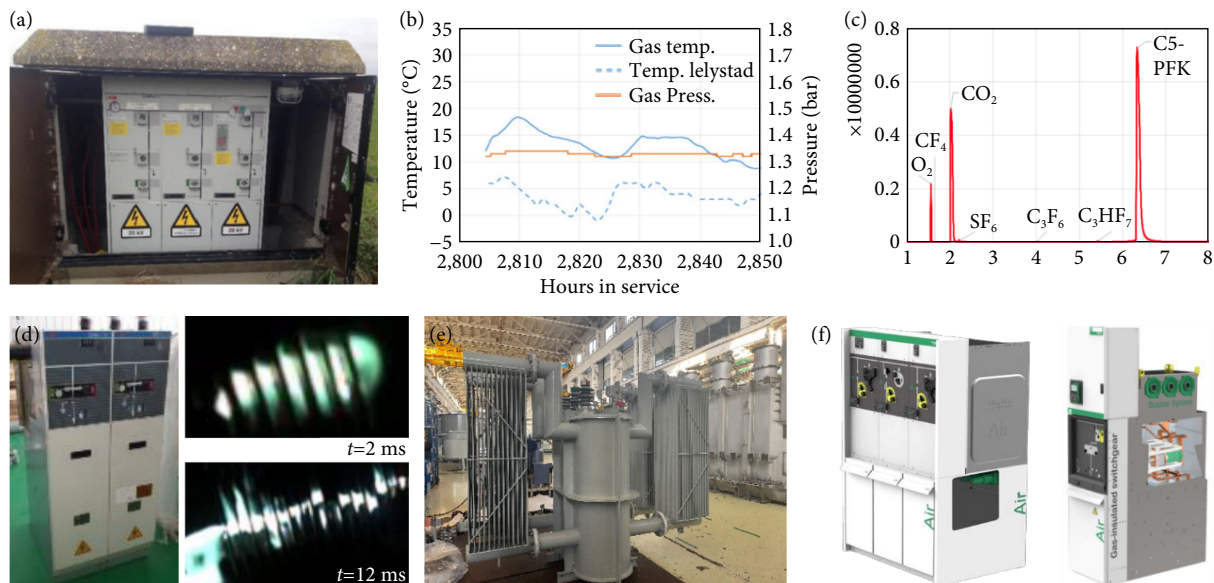
### 4.1 Medium voltage equipment

MV-GIE is widely used in an urban distribution network such as building, subway, airport. At present, several kinds of eco-friendly MV-GIE have been developed. The first C<sub>5</sub>F<sub>10</sub>O/air based ring main units (RMU) was proposed by ABB in 2015 (Figure 12(a))<sup>[226]</sup>,

which was installed as compact secondary substations (CSS) in the Netherlands and long-term field experience was obtained. The 10.7 kPa C<sub>5</sub>F<sub>10</sub>O/129.3 kPa air gas mixture achieved a service voltage of 20 kV and loads up to 630 A. The gas temperature and absolute pressure remained stable during operation, as shown in Figure 12(b). The gas composition analysis showed that heptafluoropropane (C<sub>3</sub>F<sub>7</sub>H) was the main decomposition by-product (lower than 50 μL·L<sup>-1</sup>) during the first year. The other 24 kV/630 A MV-GIS was also developed with a minimal operating temperature of -15°C. The gas sample taken from the CB that has done 100 switching operations at around 200 A had no measurable reduction of C<sub>5</sub>F<sub>10</sub>O and the by-products of C<sub>3</sub>F<sub>7</sub>H (~100 μL·L<sup>-1</sup>), CF<sub>4</sub>, C<sub>3</sub>F<sub>6</sub> were generated (Figure 12(c)). The C<sub>6</sub>F<sub>12</sub>O/CO<sub>2</sub> based 10 kV C-GIS was also proposed<sup>[58,95,227]</sup>.

The C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> based 12 kV RMU with metal plate to extinguish arc was also designed by China Electric Power Research Institute, as shown in Figure 12(d). The RMU passed the withstand voltage test of 1.2 times the rated insulation level and the active load current opening and closing tests of 630 A for 10 times and of 1000 A for 2 twice<sup>[228]</sup>. The arc in C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> demonstrated a lower voltage, wider arc channel and more liable to fall from the metal plate compared to that of SF<sub>6</sub>. The flat plate with an insulation cover and CuW static, moving contact was used to improve the switching performance and anti-ablation ability. Besides, the 10 kV/630 kVA GIT with C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> as the insulating and heat dissipation medium was developed (Figure 12(e))<sup>[228]</sup>. The average temperature rise of C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> is higher than that of SF<sub>6</sub> under the same load condition, and the internal heat dissipation structure was optimized.

Besides, the remaining uncertainties regarding the toxicity urge a preferable solution that uses natural gas or proven safe gas, such as air for dielectric medium combined with vacuum technology for current interruption in MV-GIE. Vacuum breaking is a well-known technology used for decades in MV. The moderate pressure increase of air enables SF<sub>6</sub> insulation performance and keeps the same dimensions of switchgear. Other F-gases (C<sub>5</sub>F<sub>10</sub>O or C<sub>4</sub>F<sub>7</sub>N)



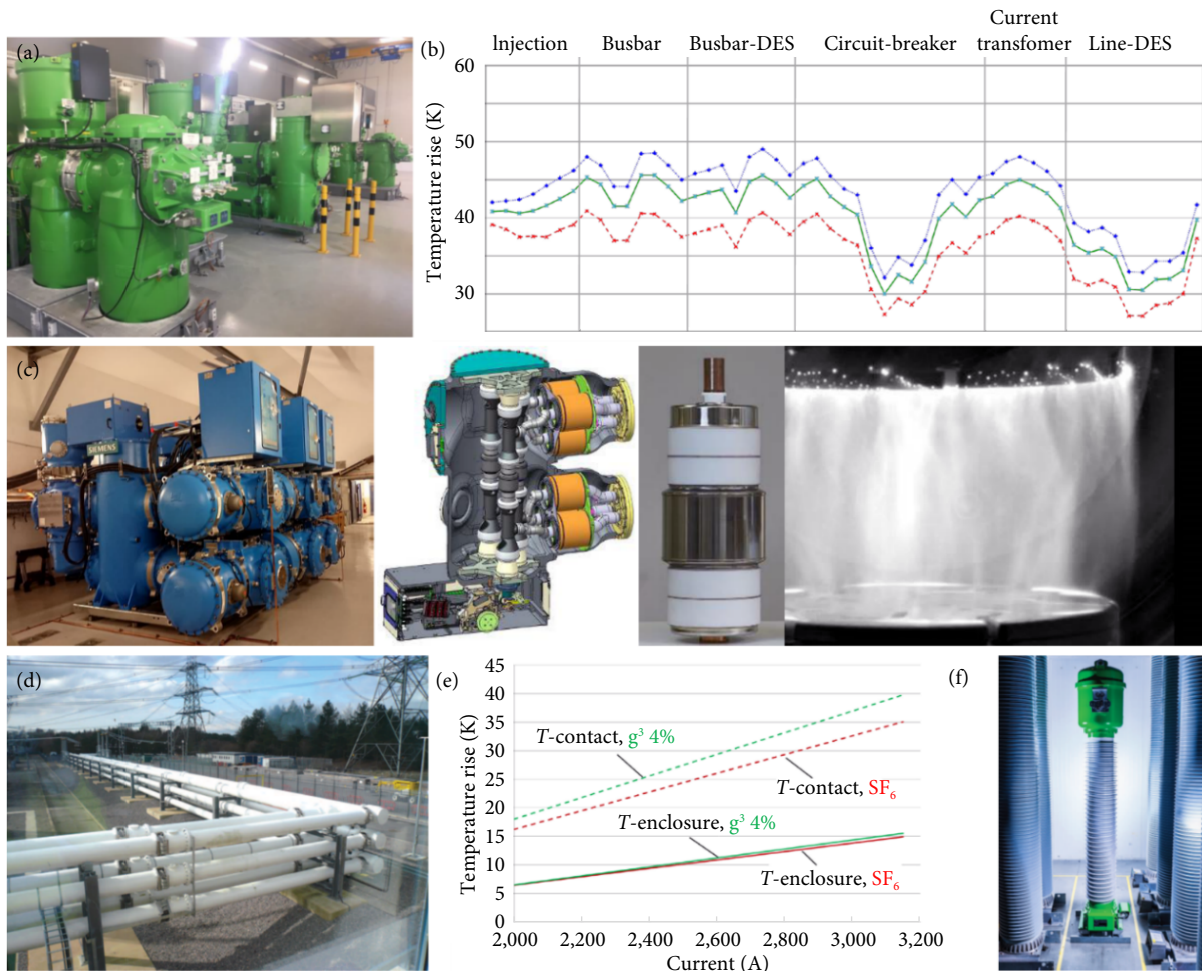
**Fig. 12** The design and application of eco-friendly insulating gas based MV-GIE. (a) Air-plus switchgear installed in the Netherlands. Reprinted with permission from Ref. [228], © 2017 IEEE. (b) Gas temperature (blue) and absolute pressure (orange) versus time recorded 2800–2850 h for the Air-plus switchgear. Reprinted with permission from Ref. [228], © 2017 IEEE. (c) Gas sample is taken from a circuit breaker that has done around 100 switching operations at around 200 A. Source: Johannes Hengstler, Maik Hyrenbach, ABB, in CIGRE WG B3.45 Meeting. (d) The 12 kV C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> Ring Main Unit. Source: Z. Li, China Electric Power Research Institute. (e) The 10 kV C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> GIT. (f) The SF<sub>6</sub>-free load break switch with shunt vacuum interruption (SVI) technology. Source: C. Preve, Schneider Electric.

with lower GWP than SF<sub>6</sub> have safety concerns, especially for MV switchgear that is close to the public. A gas container with safety concerns cannot be acceptable. The potential carcinogenic, reproductive and neurotoxic risks are not known. Moreover, these gases are per- & polyfluoroalkyl substances (PFAS). These substances are currently under the scope of Registration, Evaluation, Authorization and Restriction of Chemicals (REACH regulation of Europe) which is looking to ban all “non-essential” use. As natural alternatives such air exist, fluorocarbons may be banned in the future. Recently, Schneider Electric proposed the innovative SF<sub>6</sub>-free load break switch with shunt vacuum interruption (SVI) technology that is set in an epoxy tank (AIS) or stainless-steel tank (GIS)<sup>[229,230]</sup>. The designed 12–24 kV/630 A load break switches (LBS) keep the same operating mode as the traditional 3-position switch and have been energized in Sweden, France, China in 2019 (Figure 12(f)).

#### 4.2 High voltage equipment

The eco-friendly insulating gas based HV-GIE is also designed and applied since 2015. Specifically, GE launched the world's first C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> based 145 kV GIS with an inflation pressure of 0.85

MPa, an operating temperature of –25–40°C, and a rated current of 3150 A/40 kA (Figure 13(a))<sup>[231]</sup>. The device passed the temperature rise and insulation tests required in IEC 62271-203 standard, as well as the terminal fault (TF), short line fault (SLF, L75, and L90) and capacitor breaking test involving CBs. As the mass density of C<sub>4</sub>F<sub>7</sub>N gas mixture is lower than SF<sub>6</sub>, the gas blowing speed and pressure construction are affected during no-load operation and high current arc extinction. Therefore, the chamber volume, channel diameter, and spring of CB unit were optimized. In addition, the EPDM seal ring was also replaced and a new density meter, and pressure relief device were designed and installed. The temperature rise of the conductors given in Figure 13(b) confirmed the addition of a small portion C<sub>4</sub>F<sub>7</sub>N demonstrates a positive effect on the overall heat dissipation performance. In 2017, the first 145 kV/40 kA equipment was installed in Etzel substation in Switzerland, and operated at 123 kV at the initial stage<sup>[232]</sup>. In 2018, the same type of GIS was applied in Grimmer substation in France with an operation voltage of 72.5 kV. The maintenance strategy follows SF<sub>6</sub> related regulations and the gas mixing ratio was detected. The second generation C<sub>4</sub>F<sub>7</sub>N based 145 kV/40 kA GIS utilized O<sub>2</sub> as the second additive gas to enhance the switching



**Fig. 13** The design and application of eco-friendly insulating gas based HV-GIE. (a) The 145 kV C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub>/O<sub>2</sub> based GIS developed by GE. Source: M. Inversin, D. Signing Tsamo, RTE. (b) Temperature rise profile along the fully equipped 145 kV GIS bay at 2500 A. (Red) SF<sub>6</sub>; (Green) 6% C<sub>4</sub>F<sub>7</sub>N/94% CO<sub>2</sub> gas mixture; (Blue) CO<sub>2</sub>. Source: D. Gautschi, GE Grid Solutions. (c) The 145 kV GIS developed by Siemens with 80%N<sub>2</sub>/20%O<sub>2</sub> and vacuum interrupter(left). The vacuum interrupter with a height of 0.65 m and a diameter of 0.23 m (middle). The right shows the diffuse stable axial magnetic field (AMF) vacuum arc at 50 kA. Source: K. Kim, ILJIN Electric. (d) The 420 kV C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> GIL developed by GE. Reprinted with permission from Ref. [97], © 2016 IEEE. (e) The comparison of temperature rise between C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> and SF<sub>6</sub>. Source: D. Gautschi, GE Grid Solutions. (f) The 245 kV C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> GT developed by GE. Reprinted with permission from Ref. [97], © 2016 IEEE.

performance as O<sub>2</sub> is capable of limiting the generation of gaseous and solid by-products<sup>[233]</sup>. Besides, the HV GIS with 6% C<sub>5</sub>F<sub>10</sub>O/82% CO<sub>2</sub>/12% O<sub>2</sub> was also designed for 170 kV/40 kA scene with a working temperature of 5–40°C<sup>[234]</sup>.

The fluoride-free strategy is another solution for eco-friendly GIS, that is, using air insulation associated with vacuum breaking or CO<sub>2</sub> for insulation and breaking with pressure increase compared to SF<sub>6</sub>. For example, the world's first 145 kV/40 kA and 170 kV/50 kA GIS with clean air (80% N<sub>2</sub>/20% O<sub>2</sub>) and vacuum interrupter was developed by Siemens Energy, as shown in Figure 13(c)<sup>[235,236]</sup>. The contact geometry of CB was realized with the axial magnetic field (AMF) to ensure sufficient arc stability, strength, and suitable spatial distribution. The measured X-rays of the vacuum interrupter were far below the required thresholds of IEC and ANSI/IEEE standards. The first substation was installed and commissioned in 2019 in Norway. Besides, the design of single vacuum-interrupter units up to 245 kV and 63 kA is ongoing currently. The advances in vacuum switching technology will bring promising application for higher voltage fluoride-free GIS.

As for the other HV GIE such as GIL, GE developed the first C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> 420 kV GIL in 2016 with a working pressure of 1.06 MPa and operating temperature of –25–40°C (Figure 13(d))<sup>[96]</sup>. The equipment passed the lightning impulse, switching impulse and power frequency withstand voltage (PFVV) tests according to IEC 62271-203. The temperature rise on the enclosures with 4% C<sub>4</sub>F<sub>7</sub>N/96% CO<sub>2</sub> is similar to SF<sub>6</sub>, and the contact condition in the range of 2000–4000 A has been fully validated below the corresponding limits (Figure 13(e)). The first 420 kV/63 kA GIL was installed in 2016 at the Sellindge substation. In 2020, the world's first 1000 kV GIL was developed by Ping Gao Group, which possessed a standard unit length of 18 m, a PD intensity of less than 3 pC, and an annual leakage rate lower than 0.01%. Besides, GE also proposed the C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> based 245 kV current transformer (CT) that passed relevant special tests including PFVV, basic impulse level (BIL), PD, capacitance and dielectric factor as specified by IEC 61869 standard.

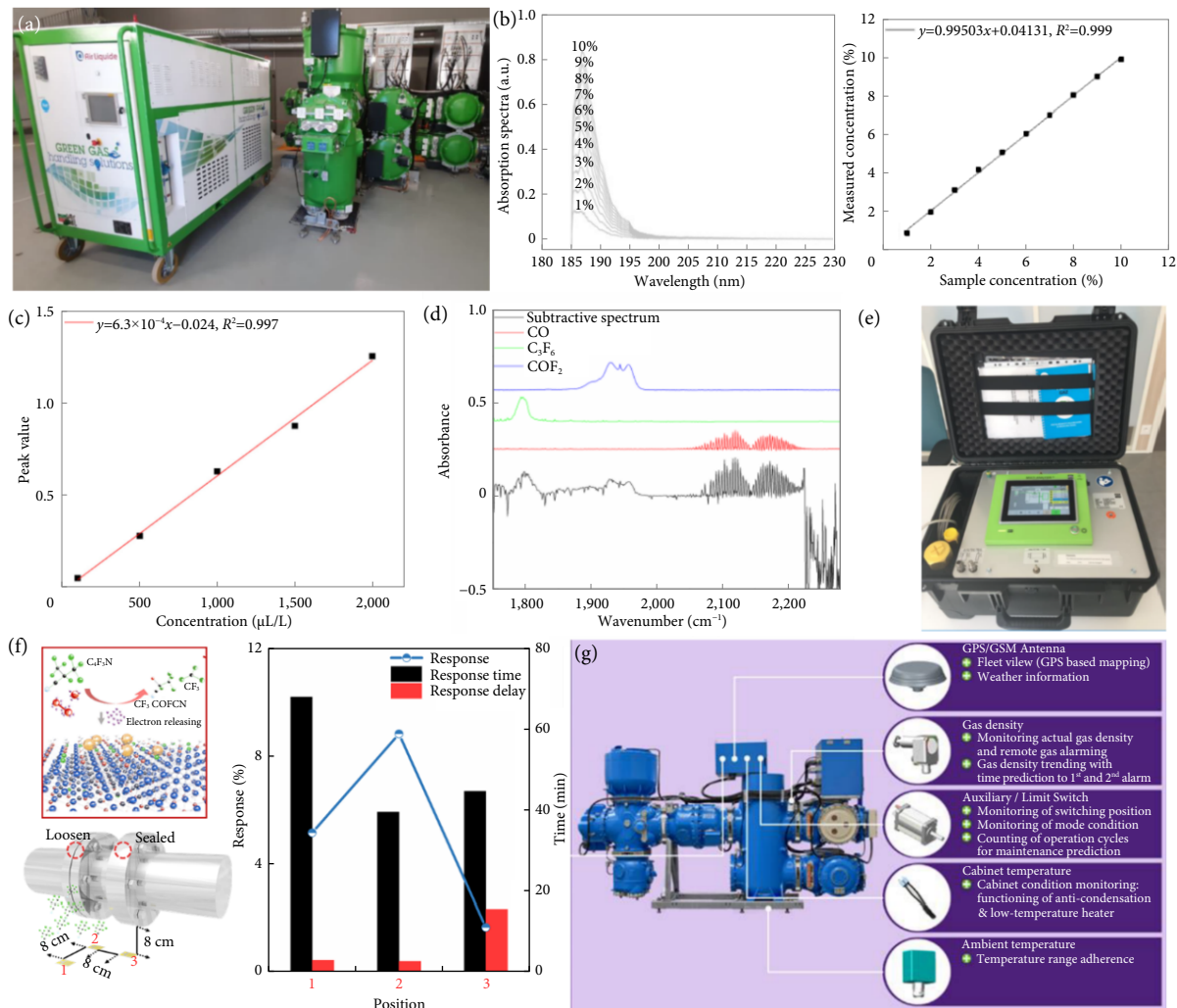
Overall, the MV and HV eco-friendly GIE developed currently offer similar performance and footprints with that of SF<sub>6</sub>. However, the heat dissipation, and current interruption capability of fluorinated gases are inferior owing to the basic physicochemical properties. The utilization of fluorinated eco-friendly gas or fluorine-free gas are the two main strategies for the next-generation SF<sub>6</sub>-free GIE, wherein the advanced possess in repeated arc-quenching of fluorinated gas and high capacity vacuum interruption technology is going to bring breakthrough in the field.

### 4.3 Maintenance related devices

The application of eco-friendly GIE also requires maintenance related devices including gas management, composition analysis and on-site monitoring, which has been developed gradually. As shown in Figure 14(a), the service cart with the gas filling, recovery and reclaim functions has been proposed and qualified by the French Transmission System Operator (TSO) with a filling accuracy less than 0.1%<sup>[231]</sup>. Frankly, there exists difference in gas detection objects between eco-friendly and traditional SF<sub>6</sub> GIE. As the main insulating gas determines the insulation level, it is necessary to timely and accurately grasp the gas mixture proportion in the equipment during operation. Meanwhile, the detection of typical or harmful decomposition by-products is also significant for eco-friendly GIE operation and fault diagnosis. The corresponding gas detection technology for eco-friendly insulating gas and relevant by-products are also investigated.

Gas absorption spectroscopy has the advantages of feasible equipment integration, high precision, and great reliability, which has promising potential for on-site gas composition analysis application. The ultraviolet, infrared spectral characteristics of eco-friendly insulating gas have been explored. For example, C<sub>4</sub>F<sub>7</sub>N possesses an obvious absorption spectrum in the ultraviolet spectral band that has great linearity with C<sub>4</sub>F<sub>7</sub>N content, as shown in Figure 14(b). The detection of C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> mixing ratio can be achieved with a detection error less than 5%<sup>[237]</sup>. The fiber-coupled LED gas sensor was developed for concentration measurement of fluorinated ketones. The dual-wavelength scheme was proposed for gas composition analysis in the harsh environment, achieving the C<sub>5</sub>F<sub>10</sub>O detection limit of  $\Delta p = 12$  Pa and relative concentration accuracy of  $\Delta c/c \approx \pm 1\%$ <sup>[238]</sup>. Ultraviolet differential optical absorption spectroscopy (UV-DOAS) was also proposed for the quantification of C<sub>5</sub>F<sub>10</sub>O, realizing the detection range of 0.1%–7.5%<sup>[239]</sup>. In view of the possible equipment gas leakage detection, the infrared-based method was proposed. As shown in Figure 14(c), the infrared band of C<sub>4</sub>F<sub>7</sub>N with higher absorption intensity and better linearity was found in the range of 750–780 cm<sup>-1</sup>. The utilization of peak area as the eigenvalue brought the detection error less than 3% for 1%–10% C<sub>4</sub>F<sub>7</sub>N. Further, the stronger infrared absorption band was found in 1240–1290 cm<sup>-1</sup>, which demonstrated a superior linear relationship with 0–0.2% C<sub>4</sub>F<sub>7</sub>N and was suitable for leakage detection<sup>[240]</sup>. Besides, the subtractive spectrum technology combined with long optical path gas cell and FTIR was employed for by-product analysis. The subtractive spectrum given in Figure 14(d) confirmed the rapid analysis of C<sub>3</sub>F<sub>6</sub>, CO, and COF<sub>2</sub> can be achieved<sup>[241]</sup>. For the device level, the gas analyzer for mixing ratio, moisture and decomposition by-products detection C<sub>4</sub>F<sub>7</sub>N has been developed (Figure 14(e))<sup>[231]</sup>. In another work, the multi-parameter sensor system was designed with a piezoresistive transmitter for pressure detection, a temperature sensor and a digital density meter for density detection. The device provides a measurement uncertainty below  $\pm 4\%$ , a resolution better than 1% of the C<sub>5</sub>F<sub>10</sub>O content and has been mounted on GIS for one year<sup>[242]</sup>.

Besides, building a novel power system with extensive interconnection and intelligent interaction features puts forward higher requirements for the intelligent perception of GIE status. Advanced micro-nano sensors with low power consumption and high sensitivity are the basic components. For eco-friendly insulating gas, the room temperature electrochemical sensor was reported recently. As illustrated in Figure 14(f), SnO<sub>2</sub> was found to possess superior sensitivity to C<sub>4</sub>F<sub>7</sub>N while a relatively high temperature is required for gas desorption<sup>[243]</sup>. The Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub>-SnO<sub>2</sub> nanocomposite with superior sensing performance was achieved, wherein Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> with specific area works as the sensing framework and SnO<sub>2</sub> nanoparticles take the role of bait to attract C<sub>4</sub>F<sub>7</sub>N molecule. The designed Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub>-SnO<sub>2</sub> gas sensor exhibited an 8.8% response to 45  $\mu\text{L}\cdot\text{L}^{-1}$  C<sub>4</sub>F<sub>7</sub>N within 180 s, which raise the sensitivity by 460% compared to the pristine Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> and provides a promising route for gas leakage monitoring<sup>[244]</sup>. Further, the “Sense-gear” concept for comprehensive monitoring of eco-friendly GIE to achieve intelligent equipment was proposed, as shown in Figure 14(g). Specifically, the GPS/GSM antenna, gas density, auxiliary/limit switch, cabinet temperature, ambient temperature sensors are embedded into the GIE during design and assemble, and relevant detected data will be collected and submitted through the Internet of Things (IoT) connectivity device. The realization of this concept will bring revolutionary progress to the SF<sub>6</sub>-free GIE in the power industry.



**Fig. 14** The design and application of maintenance related devices for eco-friendly insulating gas based GIE. (a) The gas handling equipment for C<sub>4</sub>F<sub>7</sub>N based GIE. Source: M. Inversin, D. Signing Tsamo, RTE. (b) The ultraviolet spectral absorption spectrum of the C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> gas mixture with different content of C<sub>4</sub>F<sub>7</sub>N(left) and the obtained concentration inversion curves. Reprinted with permission from Ref. [239], © 2019 Author(s). (c) The infrared spectroscopy fitting curve of C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> gas mixture with different content of C<sub>4</sub>F<sub>7</sub>N. Reprinted with permission from Ref. [242], © 2019 Elsevier Ltd. (d) The infrared spectroscopy of the C<sub>4</sub>F<sub>7</sub>N decomposition by-product C<sub>3</sub>F<sub>6</sub>, CO and COF<sub>2</sub>. Reprinted with permission from Ref. [243], © 2020 Elsevier Ltd. (e) The C<sub>4</sub>F<sub>7</sub>N gas mixture analyzer used on site. Source: M. Inversin, D. Signing Tsamo, RTE. (f) The SnO<sub>2</sub>/Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> gas sensor for C<sub>4</sub>F<sub>7</sub>N leakage detection. Reprinted with permission from Ref. [245], © 2022 American Chemical Society. (g) Overview of the Sensgear concept and measurement quantities. Source: M. Kuschel, Siemens Energy.

## 5 Challenges and perspectives

Nowadays, the increasing policy restrictions and climate control measures of SF<sub>6</sub> further promote the development and application of eco-friendly insulating gas. Although substantial efforts have been made in the field, several significant challenges remain that call for more solutions to achieve the next-generation SF<sub>6</sub>-free GIE in the future.

### 5.1 Challenges

#### Stability

The low GWP essentially reflects inferior stability, while electrical stability is highly pursued by GIE. The existing “cumulative effect” of decomposition by-products and continuous consumption of eco-friendly electron affinity gas under discharge needs to be addressed. The strategies that can hinder the critical reaction pathways of gas decomposition and product generation should be clarified. For example, the design of other “decomposition inhibi-

tion” or “by-product regulation” additive gas and optimization of operation conditions (gas pressure, mixing ratio) to improve the stability is necessary. The clarification of the limit content of various impurities in the gas mixture should also be conducted.

#### Interruption

The study on arc-quenching characteristics is significantly less extensive than that on dielectric properties. Existing research has confirmed that eco-friendly insulating gas has the load current interruption capability, while there exist challenges such as the decline of dielectric strength, and precipitation of solid by-products. Most eco-friendly gases cannot recombine into themselves after dissociation during arc extinction. This requires us to pay more attention to the arc-quenching performance of eco-friendly gases. However, some basic data, such as electron-impact cross-sections and spectrum data, are still unavailable and require more investigation. The departure from thermodynamic and chemical equilibrium in the current zero of arc extinction also brings us great difficulties in arc modeling. Further optimization on the eco-

friendly arc extinguishing composition and the structure of the CB, utilization of auxiliary arc extinguishing means, etc. to improve the interruption capacity and inhibit the solid by-product precipitation is significant to realize the reliable application in arc extinguishing scenarios.

#### Compatibility

Most eco-friendly insulating gases demonstrate great compatibility with aluminum, silver, while fluorinated-nitrile, ketones with active functional groups are incompatible with copper. This also exists for functional materials such as sealing rubber, adsorbent. The role of solid surface in the decomposition of eco-friendly insulating gas needs to be clarified, and alternative solutions or anti-corrosion strategies should be sought for incompatible materials. For example, grain boundary engineering, fluorinated coating might be useful. Besides, the highly selective adsorbent that is used for toxic, corrosive by-product elimination should be designed for specific eco-friendly insulating gas. Further investigations on the impact of additives (oxygen, etc.), and impurities on materials compatibility should be conducted.

#### Health and safety

Carcinogenic, mutagenic and reprotoxic (CMR) effects, bone marrow cytotoxic effects (cytotoxicity involves the death of cells), and neurotoxic effects of the new gas candidates must be well known in order to define the personal protection of operators and users and to insure the safety of people who could be in contact with these gases in case of accidental leakage.

#### Maintenance

The MV eco-friendly GIE has achieved small-scale application in China and various HV realized test running in the EU, etc. The operation and maintenance strategy mainly refers to the SF<sub>6</sub> GIE currently, and few standards on SF<sub>6</sub>-free GIE are proposed. The safety protection measures for eco-friendly gas and decomposition by-products with toxicity should be clarified. The monitoring of mixing ratio, gas pressure, solid precipitation, etc. is recommended for fluorocarbon based GIE to ensure long-term operation reliability. Besides, the principle for sub-package, disposal and recovery of eco-friendly insulating gas should be provided.

## 5.2 Perspectives

#### SF<sub>6</sub> control and recycling

Frankly, SF<sub>6</sub> outperforms various eco-friendly insulating gas currently in terms of stability, self-recovery, and current interruption. It is urgent to establish and improve the whole cycle tracking and supervision of SF<sub>6</sub>, and strictly prohibit random emission, to avoid the climate impact caused by SF<sub>6</sub> considering the large number of service equipment. Meanwhile, strengthening the recovery, purification, and reuse of SF<sub>6</sub> or SF<sub>6</sub>/N<sub>2</sub> to improve the reuse rate and service life of existing SF<sub>6</sub> will be a short-term trend. For the SF<sub>6</sub> waste gas that has no recycling value, the development of harmless degradation and treatment strategy to achieve in-situ disposal will be developed.

#### Insulation coordination

There exists a tradeoff between dielectric strength and liquefaction temperature of eco-friendly insulating gas, and the high sensitivity to electric field inhomogeneity also exists. Optimization of insulation design or coordination based on gas properties will further enhance the operating reliability. For example, gas-solid composite insulation design, and surface/space charge suppression needs to

be further explored. The improvement of key materials processing technology to avoid latent defects is also significant.

#### Arc-quenching mechanism

It is anticipated that the arc-quenching characteristics of eco-friendly gases will be investigated comprehensively by numerical and experimental approaches. The works on the basic data including particle compositions, thermodynamic properties, transport coefficients, radiation coefficients, and dielectric breakdown properties, will be gradually improved, with the development of new techniques, e.g. quantum chemistry and artificial intelligence. To deeper and better understand the arc interruption of eco-friendly gases, the 3-D MHD models instead of 1-D and 2-D should be developed, and more sophisticated models, such as radiative energy transport, nozzle ablation, contact erosion, turbulence, and the departure from equilibrium, should be coupled.

#### Scientific management of PFAS

In 2022, 3M decided to exit PFAS manufacturing by the end of 2025. Totally 58 kinds of fluoropolymers, fluorinated fluids, and PFAS-based additive products are affected, including C<sub>4</sub>F<sub>7</sub>N, C<sub>5</sub>F<sub>10</sub>O<sup>[245]</sup>. PFAS are critical in several fields such as semiconductors, batteries, refrigerants, airplanes, medical technologies, etc. This decision does not mean to deny the application potential of fluorinated nitrile and ketones, while the impact on application of SF<sub>6</sub>-free GIE needs to be assessed. The natural gas combined with the vacuum circuit breaker possesses technical bottlenecks in EHV and UHV application. The scientific, reasonable application and management of PFAS are appealed.

#### Development of new gas

The development of computational chemistry provides effective tools for new gas design, especially the clarification of the structure-activity relationship between gas molecules and insulation properties, environmental features, toxicity, etc. Besides, the synthesis of designed molecules also requires interdisciplinary cooperation including chemical engineering, physics, and biomedical science. The further trend is to develop non-PFAS eco-friendly insulating gas with advanced comprehensive performance.

#### Intelligent SF<sub>6</sub>-free equipment

The “novel power system” requires higher operational reliability and information perception ability of various equipment including GIE. The status assessment of eco-friendly insulating gas is also significant. The development of advanced sensing technologies and distributed devices for SF<sub>6</sub>-free GIE brings promising application reliability improvement. Meanwhile, the new gas composition sensor, discharge monitoring, and pressure detector are expected to be embedded with the equipment.

## 6 Conclusion

In this paper, we systematically summarized recent advances in eco-friendly insulating gas and the development of SF<sub>6</sub>-free GIE. The increasing SF<sub>6</sub> emission in the world urged substitutes with advanced features in dielectric insulation, arc-quenching, stability, material compatibility as well as bio-safety. The in-detail process of the design and assessment of various eco-friendly insulating gases was highlighted, and representative applications were provided. We also summarized the existing challenges and future development trends in the field, which hopefully steer the development of eco-friendly insulating gas and GIE.

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## Additional information

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## Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

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